Chapter 1
Introduction

Humans are familiar with plasmas since time immemorial: lightning, polar lights and flames are popular examples of them. Furthermore, even more than 99 % of the apparent universe is made up of plasma. But nevertheless it took until the 20th century until extensive research in this area began. This is due to the technical difficulties in performing experiments with a plasma and the mathematical complexity involved in theoretical studies. In particular even for a bottom-of-the-line plasma model one has to combine a fully developed theory of electrodynamics and classical mechanics within the framework of a many-particle-problem.

When W. Crookes noticed in 1879, that the properties of the medium in gas discharges are different from solid, liquid and gaseous materials, he called this the 'fourth state of matter' [36]. The important fact, that the charged particles in these discharges exhibits a collective behaviour due to the long ranging Coulomb forces, was already known by Lord Rayleigh in 1906 [133]. In 1929 L. Tonks and I. Langmuir [177] recognised that this state of matter can be considered as a carrier for e.g. fast-electrons, molecules and ions of gas impurities (similar to blood carrying around its various corpuscles). Thus they called this state of matter a 'plasma' [113], originating from the Greek word $\pi\lambda\alpha\sigma\mu\alpha$, meaning mouldable substance. Nevertheless plasma-physics remained a small and unimportant field in physics, until countries like Great Britain, France, the U.S.S.R. and the USA started patronising it for their political interests in nuclear weapons and power-plants at the beginning of the 1950s [18]. In particular the energy crisis in 1973 boosted the efforts in building a controlled thermonuclear fusion reactor, leading to many large scale magnetic confined fusion (MCF) experiments, like the very successful 'Joint European Torus' (JET), which started in 1983 [76]. The next step is the 'International Thermonuclear fusion Reactor', which shall demonstrate the feasibility of a commercial fusion reactor [76]. Despite these huge scale experiments plasma-physics has nowadays become indispensable even in many fields of applied physics like material science, semiconductor technology and physical chemistry.

A new field within plasma physics arose shortly after the construction of the first Laser by Maiman in 1960 [100]. As soon as people learned how to build more intense lasers with a pulse duration in the nanosecond region, scientists reported optical breakdowns in air and in solid materials [73], the first laser produced plasmas. Quite soon it was realised that lasers emitting nanosecond pulses with a pulse energy of several kilojoules are an ideal tool to study high density plasmas, the physics of nuclear weapons, thermonuclear fusion (Inertia Confinement Fusion (ICF)) and plasma instabilities [3, 35]. Consequently a number of huge nanosecond laser systems with several dozens of beams, each delivering a pulse energy beyond 1 kJ, have been built during the 1970s and 1980s: among the best known systems are PHEBUS at Limeil (France), GEKKO at Osaka (Japan), NOVA and Shiva at Livermore (USA), OMEGA at Rochester (USA) and VULCAN at the Rutherford-Appleton-Laboratories (United Kingdom) [91].

A technical milestone in laser plasma physics was accomplished with the idea of chirped pulse amplification (CPA) [101, 125, 163, 164] in 1985 by Mourou's group and the development of the first turnkey short pulse oscillator by Sibbett and his group in 1990 [155]. Soon this short pulse oscillator and the CPA amplification scheme were put together to make up the first high-power 'Table Top Terawatt' (T^3) laser [83, 166]. In contrast to the abovementioned nanosecond lasers with pulse energies usually beyond several 10 kJ, these systems often emit pulses with not much more energy than a few joules. But their pulse duration can be more than three orders of magnitude shorter, so that their peak pulse intensity even surpass the peak intensity of the nanosecond systems. The basic principle of these systems is to stretch a femtosecond pulse generated in a Kerr mode-locked Titanium-Sapphire(Ti:Sa)-oscillator to several picoseconds. In general this pulse is amplified in a series of Ti:Sa-crystals, before it is recompressed again [12, 124]. Today these T^3-systems are widely used around the world, because they are quite cheap and rather easy to operate, so that it is convenient to perform experiments with them. Among the largest operational CPA-systems at present are the VULCAN petawatt beam-line at the Rutherford-Appleton-Laboratory (United Kingdom) and the LULI petawatt beam-line at the École Polytechnique (France) with a pulse energy up to 500 J and a pulse duration between 500–1000 fs [91].

These CPA-systems also enable a new fusion scheme, called the 'fast igniter' [118, 168]. In this scheme a small sphere, made of a deuterium–tritium mixture, is heated and compressed by nanosecond laser pulses shining onto it from all sides. Then a 100 ps laser pulse drills a hole into the coronal plasma. At stagnation an additional pulse (containing several 10 kJ in 10 ps) propagates through the hole into the interior, where it produces a vast amount of energetic electrons. These electrons finally ignite the plasma. Beyond this fusion scheme many new phenomena can be explored with these CPA-systems: at intensities below 10^{15} W/cm^2 pump–probe experiments are a class of highly interesting experiments for studying ultrafast transitions in matter. An example for this kind of experiments are time resolved studies of processes like non-thermal melting and the excitation and decay of phonons [47, 136, 152]. At intensities beyond 10^{16} W/cm^2 many phenomena related to the laser–plasma interaction can be studied [58, 59]. A few examples are the emission of high order harmonics from a solid surface [13, 116, 175, 176, 182, 183, 191] or the K-α emission, due to fast electrons [59, 160]. One mechanism accelerating the electrons are plasma-waves [86]. Beyond 10^{18} W/cm^2 significant amounts of fast particles with energies of several MeV [102] are produced. These particles initiate nuclear reactions, which can produce γ-rays [53, 117], or fission the nucleus, which is particularly useful for nuclear waste disposal [93, 145]. At laser intensities above 10^{21} W/cm^2 even more phenomena, like pair production, become experimentally accessible.

Most sub-picosecond experiments involving solid targets either concentrate on studying phenomena below the plasma formation limit, or they study the laser–plasma interaction itself. In contrast to that the plasma formation process is only rarely studied [9, 60, 149, 181, 184]. This is not a serious shortcoming for metals, because they can be considered as a very cold plasma from the very beginning, due to their free electron gas. But this does not hold true for dielectric media, which initially cannot be described as a plasma. In this case the laser has to create a plasma by means of optical field ionisation at first. As this process has been rarely studied for solid materials up to today, the first part of this work concentrates on this topic. Probably the most interesting result is that the transition from an initially transparent dielectric solid to the plasma state can be used as an ultrafast switch, which allows pump–probe experiments with a temporal resolution at the order of 10 fs for radiation whose wavelengths range from the vacuum ultraviolet to the near infrared.

Soon after the initially dielectric surface is transformed to a plasma, the laser pulse starts interacting with this plasma. One phenomena occurring during this interaction is the generation of strong magnetic fields inside the plasma. Up to now only measurements of the magnetic field in the under-dense region have been performed [21, 22]. Knowledge about the fields in the over-dense region was only available through computational simulations [103, 129, 189], predicting azimuthal fields of several 100 MG. This shortcoming is addressed in the second part of this work. In this part an experiment is discussed, which observed for the first time azimuthal fields well above 400 MG. With further increasing laser intensities, these exciting results will not only be interesting from an academic point of view, but they will also be the key to studying strongly magnetised high density plasmas (such as those present at the surface of white dwarfs and neutron stars) in the laboratory for the first time.

Chapter 2
Ultrafast Ionisation and Optical Gating

2.1 Introduction

As already mentioned beforehand, only a few experiments [9, 60, 149, 181, 184] focus onto the formation of a solid density like plasma from a dielectric surface, i.e. the transition from a cold solid material to a hot high density plasma. In addition, despite a publication from P. Blanc [19], there are no detailed experimental studies of plasma formation at the surface of a solid on a timescale well below 100 fs to the best knowledge of the author. Furthermore computer codes, used to simulate laser plasma interactions at solid densities, require a pre-ionised initial state in general, because ionisation models are only implemented to a very limited extent [17, 44, 45, 86, 132]. An exception are specialised codes, such as those developed by Ruhl and Mulser [137]. Furthermore it is remarkable, that there are not much more detailed studies of this process within an ultrashort timeframe, because this process has been proposed to be used for pre-pulse removal from ultrashort laser pulses. This application is known under the term *plasma mirror* in the scientific literature [61, 77, 142, 170, 173, 181]. In addition it is rarely discussed, that the phase transition from the solid state to a high density plasma can be used as a transmission gate as well. In Michelmann et. al. [109, 110] this gate is applied to characterise a 248 nm, 500 fs pulse. In the theoretical part of [110] we briefly showed (a more elaborate discussion is given in [174]), that the response time of this gate is not much longer than 50 fs. Beyond these studies it is interesting to explore, if the plasma switch is also applicable to pulses well below 100 fs or if the measured pulse properties are influenced by the increasing electron density. In this case one can try to derive the plasma properties from the measured pulse properties. These studies are the main scope of the present chapter. In its first part this issue is approached from a theoretical point of view by developing a macroscopic model describing the optical properties during the plasma formation process at a dielectric surface [174]. Based on this model, not only the optical properties of the transmission gate are calculated, but also valuable insight into the feasibility of the plasma mirror for pulse cleaning on ultrashort timescales is obtained. The experimental part of this chapter concentrates on the transmission properties of the emerging plasma, which are finally compared to the theoretical findings.

Even if the time resolution of measuring instruments increased quite a lot during the last few decades, femtosecond time resolution is still achieved by measuring the correlation between two signals. A widespread class of these correlation measurements are *pump–probe experiments* [134], which have been used successfully during the last years to study ultrafast melting, changing absorption, reflectivity and transmission of transparent semiconductors and insulators (see, e.g. [9, 47, 136, 149, 152, 180]) on a picosecond timescale. Even more recently, time-resolved x-ray reflectivity measurements have become a standard tool for studying phase transitions in solid state physics on time-scales below 1 ps [47].

In our experiment the main laser pulse is split into a strong *pump pulse*, producing the plasma, and a much weaker *probe pulse*. Both pulses are focused onto the same area of an initially transparent target. As the probe pulse can be delayed in time with respect to the pump pulse, one can measure the correlation between the probe pulse and the optical properties of the plasma, respectively. This correlation can be used either for studying the time-dependent optical properties of the plasma, if the properties of the probe pulse are known, or for measuring the properties of the probe pulse, if the time dependent optical properties of the plasma are known.

2.2 A Time-Dependent Model of Optical Plasma Properties

Concerning the plasma formation process one can distinguish two kinds of solid materials: metals and dielectrics [138]. The free electron gas present in a metallic solid is already a plasma with an electron temperature of some meV. Thus the electric field of the laser can directly interact with the free electrons of this plasma, leading to collisional absorption, also called inverse bremsstrahlung [73, 172]. This heats up the initially cold plasma and increases the electron density even further by collisional ionisation. On the other hand, in a dielectric solid there are practically no free electrons present, so that this mechanism does not work initially. Instead of this, optical field ionisation (OFI) [50] initiates the ionisation process. As soon as the increasing electron density surpasses the so called critical electron density n_{ec}, an initially transparent dielectric material becomes opaque at its surface. n_{ec} is given by

$$n_{ec} = \frac{\epsilon_0 m_e}{e^2}\,\omega_0^2 \tag{2.1}$$

with the dielectric constant ϵ_0 in vacuum, the electron charge e, the electron mass m_e and the angular frequency ω_0 of the laser field. A schematic overview of our macroscopic plasma model describing this transition process is shown in Figure 2.1.

2.2.1 The Ionisation Process

To obtain a more detailed understanding of OFI it is useful to introduce the so called Keldysh parameter $\Gamma := \sqrt{\frac{I_{ioni}}{2U_q}}$, whereby I_{ioni} is the ionisation potential and U_q is the quiver energy:

$$U_q := \frac{e^2}{4m_e}\sqrt{\frac{\mu_0}{\epsilon_0}}\,\frac{I_{pump}}{\omega_0^2} \tag{2.2}$$

with the laser intensity I_{pump}, the dielectric constant ϵ_0 and the magnetic permeability μ_0. Then one can distinguish three different domains of OFI: a low intensity domain with ionisation mainly due to multi-photon absorption ($\Gamma \gg 1$), a high intensity domain ($\Gamma \ll 1$) with tunnel ionisation and a transition region. In all three domains the OFI is described fairly accurately with the Ammosov-Delone-Krainov(ADK) theory [5], which

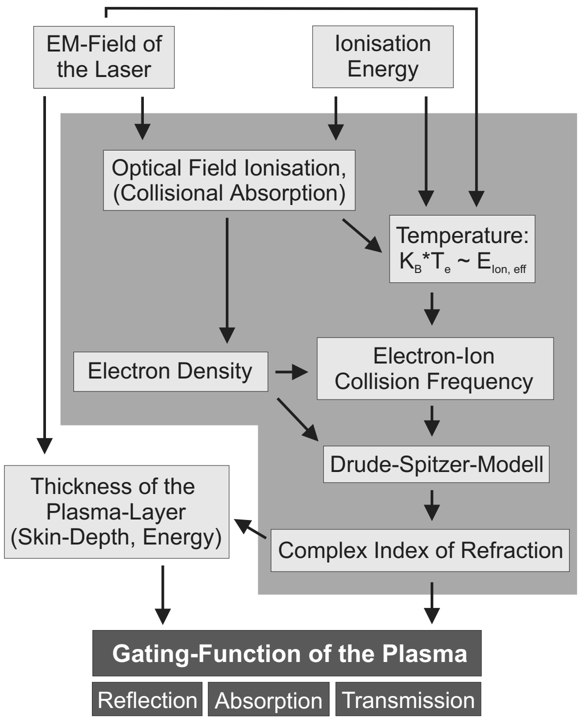

Figure 2.1: Physical model to calculate the time-dependent transmission properties of a plasma produced at an initially dielectric and transparent solid in the early phase of plasma formation.

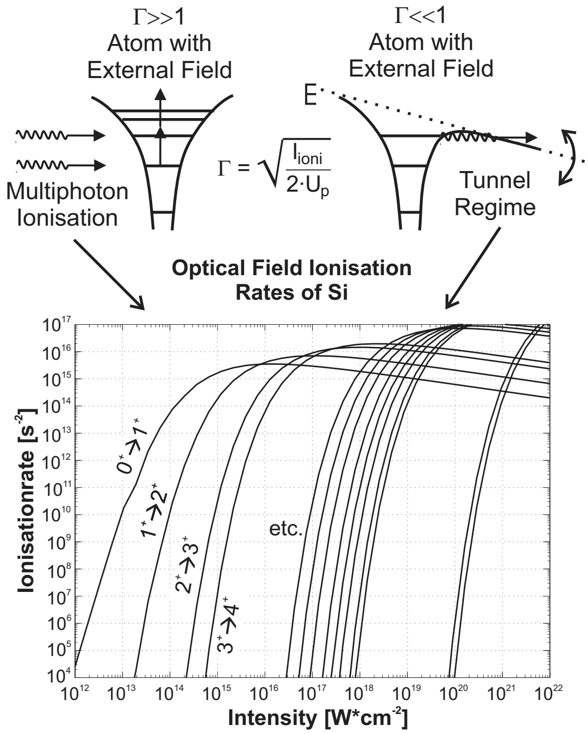

Figure 2.2: Optical field ionisation rates of Silicon from the multi-photon to the tunnel ionisation regime.

is an elaboration of the Keldysh theory [78], based on the Schrödinger equation and a scattering matrix approach. The latter theory is used in our plasma formation model (see Figure 2.1). In some cases this theory underestimates the ionisation rates as a comparison between calculated rates and experimental data shows [8, 50]. More serious are the restrictions of Keldysh's and similar theories [55] to a single atom. Extending these theories to a high density solid is a very extensive task, as one has to use a detailed quantum mechanical model of the solid. In the end, one would derive even higher ionisation rates, because the band structure of the solid almost always allows resonant absorption of photons [78]. Consequently this increases the rapidity of the switch even more. In summary, the Keldysh model is a fairly simple, but sufficiently sophisticated model to estimate lower boundaries for the ionisation rates during the plasma formation on the surface of a solid, if the ionisation potential, the incident laser frequency and intensity are known (see Figure 2.2). From these calculated rates, the time-dependent electron density n_e can be calculated easily by summing up the different transitions and taking the occupation of states into account (see Figure 2.3). It is noteworthy, that we obtained similar results, using the barrier suppression ionisation (BSI) model [7, 8].

The free electrons produced by OFI immediately gain a quiver energy up to U_q from the electric field of the laser, so that they can contribute to the ionisation rate by collisional ionisation. Consequently a binary encounter approximation of collisional ionisation, developed by Gryzinski [67], was included in the calculations. As can be seen from Figure 2.3, this only increases the electron density n_e significantly at times, when n_e is

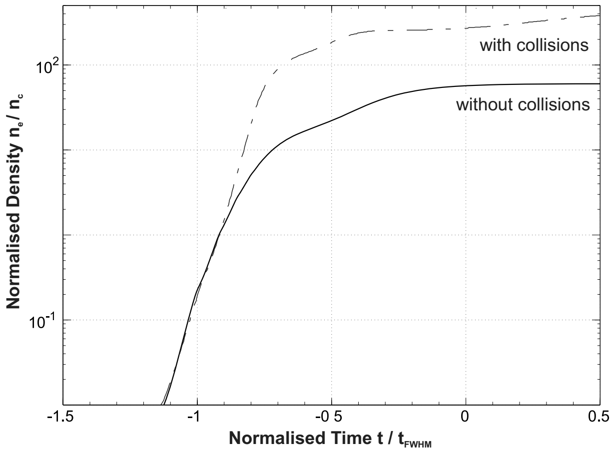

Figure 2.3: Time-dependent electron density with and without collisional ionisation. The time is normalised to t_{FWHM}, i.e. the FWHM of the pump pulse (800 nm, 60 fs, 10^{15} W/cm^2). Zero is at the peak intensity of the pump pulse.

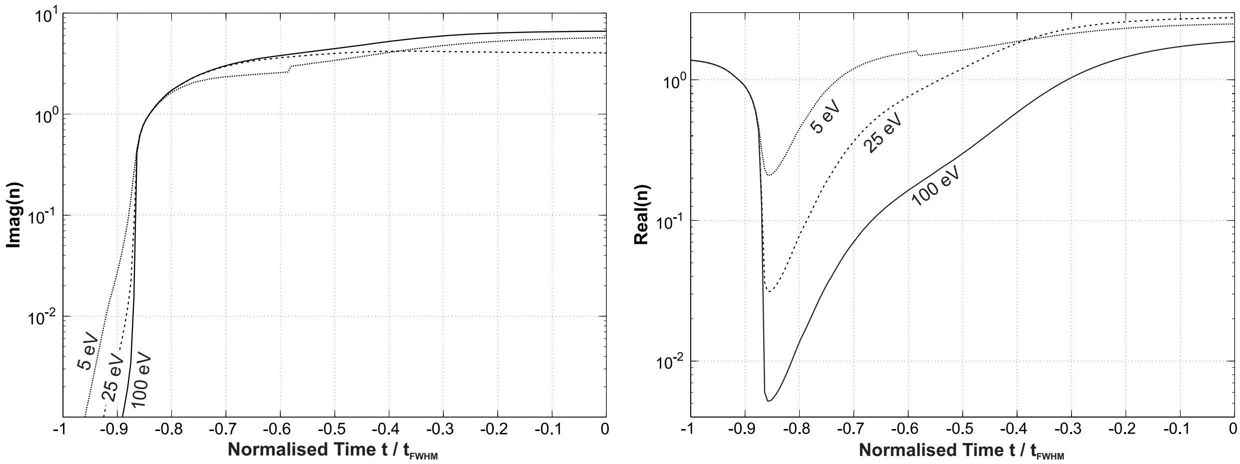

Figure 2.4: Complex index of refraction versus time for constant electron temperatures at the surface of a fused silica plate. The time is normalised to the FWHM of the pump pulse (800 nm, 60 fs, 10^{15} W/cm^2), zero is at the peak intensity of the pulse.

at least approximately as large as n_{ec}. A closer look shows that collisional ionisation slightly reduces the transition time and shifts the transition point (i.e. the time when $n_e(t)$ reaches n_{ec}) by a few femtoseconds. Thus, in summary, the transmission properties are not changed substantially by collisional ionisation.

2.2.2 The Time-Dependent Optical Properties

The next major step is to calculate the time-dependent optical properties of a thin plasma sheet, i.e. its complex index of refraction n. This can be accomplished using the Drude's model [24] of a metallic surface:

$$n^2 = \epsilon_\infty - \frac{1}{1+f^2}\frac{n_e}{n_{ec}} + i\frac{f}{1+f^2}\frac{n_e}{n_{ec}} \tag{2.3}$$

where ϵ_∞ is the dielectric function [187] at infinite frequency and f is a damping factor, equal to the electron–ion collision frequency normalised to the angular frequency ω_0 of the laser pulse. This damping factor can be estimated from Spitzer's theory [156]:

$$f = \frac{4\sqrt{2\pi}\,e^4}{(4\pi\epsilon_0)^2\sqrt{m_e}}\frac{n_e}{n_{ec}}\frac{Z_{eff}^2}{T_e^{3/2}}\frac{\ln\Lambda}{\omega_0} \tag{2.4}$$

In the last equation Z_{eff} is the average degree of ionisation, $\ln\Lambda$ is the Coulomb logarithm, m_e is the electron mass and T_e is the electron temperature. Because of the quickly increasing ionisation the plasma is far from thermal equilibrium, thus, strictly speaking, a temperature is not defined. However, if the laser intensity is weak, then a reasonably good estimate of T_e is provided by the ionisation potential of the electrons, because one can argue that the electrons need a kinetic energy of the same order as the ionisation potential to prevent immediate recombination [114]. With increasing laser intensity the cycle averaged quiver energy [130], that the electrons gain in the oscillating laser field, becomes much larger than the ionisation potential, so that an estimate of T_e is given by the quiver energy. Note that the ADK theory limits the initial electron energy to 0.2 % of the quiver energy [5, 108]. Furthermore it works out that the temporal evolution of the transmission, derived from $\Im(n(t))$, depends only weakly on T_e (left plot in Figure 2.4). Note that this does not hold for the reflection, derived from $\Re(n(t))$ (right plot in Figure 2.4). Thus for modelling the transmission through the plasma a detailed knowledge of T_e is of minor importance, whilst it is important for calculating the reflection.

To calculate the transmissivity and reflectivity of the whole plasma, we consider a thin homogeneous plasma layer with a thickness dx first. The transmission through this layer is given by [24]:

$$T(t) = \exp\left(-\alpha(x,t)\,dx\right) \tag{2.5}$$

with the absorption coefficient $\alpha(t)$ and the speed of light c_0:

$$\alpha(t) = 2\frac{\omega_0}{c_0}\Im(n(t)) \tag{2.6}$$

On the other hand the reflectivity is given by Fresnel's equation [24], i.e. in the case of normal incidence:

$$R(t) = \frac{(\Re(n(t)) - 1)^2 + (\Im(n(t)))^2}{(\Re(n(t)) + 1)^2 + (\Im(n(t)))^2} \tag{2.7}$$

Hence the total transmission of the plasma layer is:

$$T_{tot}(t) \approx (1 - R(t))\; T(t) \tag{2.8}$$

The reciprocal of Eq. (2.6) is called the skin depth $l_{skin}(t)$ and is equivalent to the length over which the intensity of the radiation propagating through the plasma is absorbed by the free electron-gas to $1/e$ of its initial value. This absorption mechanism is known as the skin effect, too.

In addition we define the ionisation depth:

$$l_{ioni}(t) := \frac{W_{n_e}(t)}{I_{pump}(t)} \tag{2.9}$$

where $W_{n_e}(t)$ is the power per volume, necessary to increase the ionisation according to the Keldysh ionisation rates, and $I_{pump}(t)$ is the pump pulse intensity. Thus $l_{ioni}(t)$ is an estimate of the plasma layer thickness, which can be ionised further, if the instantaneous power of the laser is fully consumed by the ionisation.

As shown in Figure 2.5 for early times $l_{ioni}(t)$ is much smaller than $l_{skin}(t)$, so that the absorption of the probe pulse in the plasma can be neglected. In this case mainly the intensity of the pump pulse limits the thickness of the ionised plasma layer. If the plasma is almost opaque, then this situation starts turning upside down and $l_{ioni}(t) \gg l_{skin}(t)$ begins to hold. Now the thickness of the plasma layer with an increasing ionisation is determined by the skin depth. Consequently one can use the ionisation depth $l_{ioni}(t)$ and the skin depth $l_{skin}(t)$, whichever is smaller, to estimate the time dependent thickness of the plasma layer with rapidly growing ionisation. The ionisation process becomes more and more limited to a thinner layer at the target surface, because $l_{ioni}(t)$ and $l_{skin}(t)$ decrease with time. As one can neglect the hydrodynamic expansion of the plasma on these ultrashort time-scales, it is straight forward to derive an electron density profile along the axis of the incident laser beam. This profile has its peak density at the target surface and steeply falls off into the bulk. By numerical integration of Eq. (2.5) and Eq. (2.7) over this density profile one finally derives the reflectivity and transmissivity of the plasma.

2.2.3 Results and Validity of the Model

As already discussed in Subsection 2.2.1 on a longer time-scale collisional ionisation increases the ionisation rates considerable. Furthermore, even at very early times processes like multiple ionisation [46] and resonant multi-photon absorption [73] can increase the rates beyond the values calculated with the Keldysh theory. Anyway, all of them increase the rapidity of the switch even more and mainly advance the transition point by a few femtoseconds with respect to the pump pulse maximum. It is important to note, that other seemingly important processes can be neglected on time scales below hundred femtoseconds. Among them are the heating and ionisation of regions further away from the target surface, as this occurs on the longer time scale of the non-linear heat wave propagation. Using even an optimistic value of its propagation velocity of much more than 10^7 cm/s one easily calculates, that the duration for heating a depth of 1 µm requires at least several hundred femtoseconds. Also heating by highly energetic electrons [86] is of minor importance, because this requires higher laser intensities than the only moderate intensity of up to 10^{15} W/cm^2.

As shown in Figure 2.3 the electron density increases nearly exponentially with time, which is almost independent of the material used within the calculations. Thus the particular ionisation potentials of the material are of minor importance and the exponential increase is mainly determined by the laser pulse itself.

In Subsection 2.2.2 it was already discussed, that our model describes the reflectivity $R(t)$ less precisely than the transmissivity. Even worse for calculating $R(t)$ one has to derive the dielectric constant ϵ_∞ [187] included in $\Re(n(t))$ (see Eq. (2.3)) in addition. This constant is strongly dependent on the structure of matter, which changes drastically during the ionisation process. Thus our assumption, that ϵ_∞ is constant, is a rough description of the physical reality only. Nevertheless this coarse model gives valuable insight into the reflectivity of the plasma surface: initially there is a low but constant