CHAPTER 1

Introduction

1.1. Laser plasma interactions

The main aim of the work reported in the present thesis is the experimental and theoretical studies of the fundamental physical phenomena related to laser pulse interaction with dielectric or metallic surfaces. High power laser-matter interaction results in the laser ablation and laser induced plasma (LIP) formations. The ablation and plasma properties are directly related to the physical mechanisms of laser pulse energy absorption and its redistribution in the solid and laser plasma. Studies on plasma have applications in diverse areas of science and technology. For example, in an effort to harness fusion energy on earth, physicists study devices that create and confine very hot plasmas in magnetic fields. In space, plasma processes are largely responsible for shielding earth from cosmic radiation, and much of the sun's influence on earth occurs by energy transfer through the ionized layers of the upper atmosphere. Plasma dynamics is the key field of study in understanding stellar structures.

Studies in the field of high intensity laser interaction with matter unfold numerous exotic phenomena so that the subject always stands among the current topics of research. Progress in short pulse laser technology has been in fast pace since the invention of chirped pulse amplification in 1985. At present, pulses shorter than an atto second are available. Today, Peta watt (PW) lasers are already in operation and Exa watt (EW) lasers have been successfully installed in Europe [1]. Ordinary matter will easily get ionized when subjected to high intensity irradiation. The electrons released by ionization processes are then immediately caught in the laser field and oscillate with a characteristic energy dictated by the subsequent laws of interaction. The four parameters governing the laser radiation during the interaction are wavelength, energy, pulse duration and focal spot size.

During the initial evolution of LIP, one can roughly assume a one-dimensional cartesian expansion as long as the expansion distance is much smaller than the focal spot diameter. When the plasma expands in ambient air, the plasma expansion at later times

can be reasonably taken to be hemispherical. Several computational models [2-7] have been proposed to explain the physical phenomena involved in optical breakdown of solid materials. Modelling of optical breakdown are carried out, based on the effects of avalanche ionization, electron – ion recombination, electron diffusion and so on. Niemz [8] found a square root dependence of the energy density on the pulse duration in the pico-second and nano-second range. Nano-second time scale pulses can interact with the expanding plasma so that the electron density, temperature of the excited species and so on, are sustained longer than those due to ultra-short laser pulses.

1.2. Plasma hydrodynamics and numerical modelling

Plasma theory is a combination of electromagnetic theory and theory of hydrodynamics related to plasma. There are various hydrodynamic descriptions of plasma starting with the one fluid model of Alven. General macroscopic equations for fully ionized plasma are the two fluid equations derived by Schluter and Biermann [9]. The different physical models for LIP incorporate the salient plasma features, laser structure, and numerical algorithms that accurately solve the governing equations. The whole processes of target ablation, implosion and ignition are essentially determined by hydrodynamics.

In the case of fluids, there is a velocity field whose temporal derivative corresponds to the acceleration, a mass density field $\rho(x, y, z, t)$ which corresponds to the mass m and a force density which is given by the gradient of the pressure field $p(x, y, z, t)$. Consider the fluid as being composed of electrons of mass m, density n_e, temperature T_e and ions of mass m_i, density n_i and charge Z. Assuming space charge neutrality, $n_i = n_e / Z$, the mass density field is given by,

$$\rho(x, y, z, t) = m_i n_i(x, y, z, t) + m n_e(x, y, z, t) \tag{1.1}$$

and the pressure field is,

$$p(x, y, z, t) = n_e K T_e + n_i K T_i \approx (1 + Z) n_i K T_e \tag{1.2}$$

The first basic equation is Euler equation which corresponds to the conservation of momentum:

$$\rho \frac{\partial \mathrm{v}}{\partial t} + \rho \mathrm{v} \cdot \nabla \mathrm{v} = -\nabla \mathrm{p} \tag{1.3}$$

The second basic equation is the conservation of mass, which is the equation of continuity:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathrm{v}) = 0 \tag{1.4}$$

In addition to these two, the equation of energy conservation is needed to arrive at the complete set of differential equations for uniquely solving the gas dynamic expansion of LIP. This equation is of the type,

$$\frac{\partial}{\partial t}\frac{\rho}{2}\mathrm{v}^2 = -\frac{\partial}{\partial t} n_i KT(1+Z) - \nabla \cdot (\kappa_T \nabla T) + W \tag{1.5}$$

where, LHS describes the temporal change of the kinetic energy of the fluid to be compensated by the change of internal energy (first term on RHS), by thermal conduction, characterized by the thermal conductivity κ_T and by power density W of radiation. In LIP, the net electrodynamic potentials can change in time. These components will then have to be included as additional potentials [10].

Fluid equations for the conservation of mass, momentum and energy are solved by means of suitable codes. The plasma is treated as a single fluid described mainly by a set of four main variables: density, velocity, electron, and ion temperature, which are functions of spatial coordinates and time. The fluid velocity is obtained by solving the momentum equation, using the appropriate pressure boundary condition at the interface with the ambient air. The plasma temperature can be obtained by solving the electron and ion energy conservation equations in which the source terms include the absorption of the laser energy, the heat conduction, the electron–ion coupling and the radiation loss term. The radiative energy losses, local energy changes due to radiative transfer within the plasma and radiative losses through the boundaries are also to be taken into account [11-14].

For moderate background pressures, the hypothesis of a spherical expansion appears to be reasonable for the description of near-axis plume behavior [15]. However, in high vacuum, the plume is essentially forward directed, and therefore a one-dimensional fluid dynamic model can be used to describe the system [16-22]. This model considers only the stages of expansion during and after the laser pulse without detailing the evaporation process. At the end of the laser pulse, a plume is assumed to be formed in the direction perpendicular to the target, in the same position of the irradiated spot center. The laser energy is spent on melting, vaporization and heating of the target material and

on heating and ionization of the vaporized particles. Before the expansion, the plasma can be considered to be in thermal equilibrium. After pulse termination, the cloud begins to expand perpendicular to the target into the ambient gas at specific pressure. The plasma has been treated as a single fluid characterized by one velocity and one temperature. This fluid dynamic approach disregards the effects of space-charge separation, as the time-averaged electric field of the laser radiation is zero. In fact, numerous studies have shown that the angular distribution of the laser-generated flux is often much more strongly forward peaked than the flux obtainable from small-area effusive sources operating under collision-less conditions. This forward peaking phenomenon for deposition in vacuum is now generally accepted as arising from collisions of the plume species among themselves.

Quite different approaches to simulate the LIP expansion have been proposed by Wood et al. [23, 24]. The Wood's model is based on a combination of multiple scattering and hydrodynamic approaches. The plume is allowed to be broken into different orders of scattering, whose particles can undergo many collisions with the background. Particles can only be transferred from one order to the next higher order by collisions. The densities in the individual orders propagate according to the usual conservation equations to give the overall plume expansion. Nemirovsky et al. [25], starting from the Boltzmann equation, have derived the hydrodynamic equations of motion for partially ionized plasma when the charged component and the neutral component have different flow velocities and temperatures. They have developed a general approach for the hydrodynamics of a gas in a binary mixture, when the interaction between particles of the same species is much stronger than that between particles of different species.

Le et al. [26] demonstrates a persistent lack of equilibrium between the electron and heavy particles in the expanding plasma plume. To take this effect into account, the fluid dynamic method is coupled with a kinetic approach. The processes included in the model are the ionization of the ground state and the three-body recombination as well as the photo-recombination into the ground state.

To overcome the limits of the gas-dynamic approach, many authors have investigated the plume expansion and the effects of collisions amongst particles desorbed from solid surfaces by means of direct Monte Carlo simulation. According to such a simulation by Urbassek et al [27], light and heavy particles are spatially segregated due to

the different velocity of desorbed heavy and light particles. In the back part of the cloud both species appear to be well mixed while the front part of the cloud consists mainly of light species. Itina et al [28] showed that at low pressure (0.01 mTorr), heavy particles are more energetic than light particles and their distribution of mean energy is more focused towards the center. As the background pressure increases, the mean energies of both species diminish and the distributions become less focused toward the center. This effect is more evident for light particles than for heavy particles. The decrease of the kinetic energy of the plume particles is due to the collisions with the background gas, which thermalize the particles. The decline of energy is more pronounced for light species, because they lose energy more efficiently in a collision. Therefore a smaller background pressure is required for thermalization of light components. At high pressures (100 mTorr) most of the particles are thermalized and energy distributions become broadened along the normal of the surface. The difficulties of the gas-dynamic description of the diffusive processes are avoided in such models.

To simulate the real system under study, it is necessary to include chemical models also into the fluid dynamic code. For example, in a TiO plasma in local thermodynamic equilibrium (LTE), the plasma thermodynamics is completely defined by two independent parameters, such as pressure and temperature, or enthalpy and pressure. A strong non-linear coupling occurs between the chemical kinetics and the fluid dynamics [17].

A fluid dynamic model to investigate the effects of finite rate coefficients on the plume expansion for a titanium plasma includes (a) ionization by electron impact (b) three-body recombination and (c) radiative recombination [16, 29, 19]. The simulations reveal the strong influence of plasma chemical processes on the time of flight (TOF) plots. The plasma, initially produced by the laser–matter interaction is supposed to be completely ionized. When kinetic processes of plasma chemical reactions are included, the TOF plots of Ti become significant, due to recombination processes. At the first instant, the plume arriving at 0.5 mm is essentially composed of ions, as the kinetic effect is still negligible. As time increases, recombination takes place, and consequently, the Ti molar fraction grows up, while the ion molar fraction decreases. TOF plot analysis highlights an apparent separation between atom and ion concentrations, when kinetics is introduced in the numerical modelling [29].

1.3. Outcomes of numerical modelling

The results of modelling suggest that plasma properties depend more strongly on the fluence near the ablation threshold. However, for the higher fluences of interest, the temperature and electron density reach a saturation value due to the increase of the plasma radiative cooling with the plasma temperature. Ablation of metals occurs at relatively low intensity compared with that for a transparent dielectric whose absorption is negligibly small resulting in large threshold for optical breakdown. For typical dielectrics like ceramics, energies involved in ablation are close to those required for heating the targets beyond their melting temperatures and initiate significant evaporation. Material ejection is accompanied by the formation of a vapor or plasma just above the target surface and an easily recognizable snapping sound as the velocity of some of the species exceeds the speed of sound in the immediate environment. In order to make the process efficient, so that energy is not lost due to carrier or thermal diffusion during the laser light absorption, short laser pulses should be used at a wavelength strongly absorbed by the material. For power densities above the vaporization threshold, the dominant mechanism is vaporization [30, 31].

There exist two zones in the model described by Aden et al [15], at pressures below 1 mbar: one which is directly attached to the target surface throughout the whole process, and the second is recognized as an outward moving shock front. Heavier background gases, such as *Ar* result in a slower expansion component of the vapour plume and hence lower plume velocity and shorter plume length. The plasma temperature and ionization degree seem not to be affected by the background gas, up to 100 ns. The background gases with lower ionization potential exhibit a higher ionization degree in the plasma. Further, the threshold for plasma formation in the background gas is determined by the ionization potential of the gas. Bogaerts et al [32] predicts that most of the laser–plasma interaction occurs with the background vapour, so that plasma shielding (change in the efficiency of laser energy coupling to the target by increased absorption and/or reflection from the laser-induced plasma) is only a little bit more pronounced in the gases with lower ionization potential, such as Ar, than in He.

A three dimensional Monte Carlo simulation of LIP in vacuum indicates that, when more than a few monolayers are ablated, the laser energy absorption by the evaporated particles has dominant effects on the plume shape during the expansion

process. A fraction of the recombination of ionic and excited species leads to a delayed kinetic energy transfer in the plume. This has a significant effect on the angular and kinetic energy distributions of the evaporated particles [33].

1.4. Plasma emissions induced by nanosecond and femtosecond laser pulses

In typical nanosecond laser ablation processes, two types of photon absorption mechanisms dominate. The first one is inverse bremsstrahlung absorption, which involves photon absorption by free electrons. Free electrons can gain energy from the laser beam through collisions with neutrals and ions. The probability of photon absorption due to electron-neutral collisions is much smaller than that due to electron-ion collisions. The second mechanism is photo-ionization of ground- and/or excited-state species, and multi-photon ionization for sufficiently high laser intensity. A bound electron is excited to the free energy level and thus ionized by absorbing one or more photons. The effect of the inverse bremsstrahlung process becomes more important for longer wavelengths, whereas the opposite is usually true for photo-ionization and multi-photon ionization processes [34].

Femtosecond laser beams can induce properties, which might allow improvements in analytical treatment of current nanosecond-laser-induced breakdown spectroscopy. Femtosecond laser interactions with matter have the potential for innovative materials applications. Clean craters were observed by interferometric microscope measurements [35] indicating the advantages and potential for applying femtosecond lasers to micromachining and advanced materials treatment. Due to a much smaller thermal diffusion depth, high-precision ablation and minimal damage can be obtained with femtosecond lasers. Femtosecond regime is better than the picosecond and the nanosecond ones for precise material processing because of full vaporization of the matter and no trace of molten material [36]. This regime occurs at the laser pulse duration less than 200 fs. For femtosecond laser-material interactions, only a very small fraction of the laser pulse energy is transmitted as heat and transferred to the material surrounding the laser-irradiated area. Consequently, femtosecond laser pulses can induce non-thermal structural changes, driven directly by electronic excitation and associated nonlinear processes, before the material lattice has equilibrated with the excited carriers. This fast

mode of material modification can result in vanishing thermal stress and minimal collateral damage for processing practically any solid-state material.

An intermediate regime takes place at the laser pulse duration between 0.5 ps and 100 ps. The most appropriate description of the heating and expansion processes in this regime is given by the conventional two-temperature approach [37-40].

At the longer laser pulse duration (~ ns), the heat conduction and hydrodynamic motion dominate the ablation process. The electrons and lattice ions are in equilibrium early in the beginning of the laser pulse. Hence, the limiting case of thermal ablation is suitable for the description of the long pulse ablation mode. The total melt and evaporation depth increase slightly for longer laser pulses, because the target is exposed to the laser for a longer time, and it is found that the plasma shielding is a bit less pronounced for longer pulses, because of the lower irradiance, so that the net laser fluence reaching the target increases slightly with pulse duration [41].

For energies higher than 3 mJ, there is little difference between 50 ps and 10 ns pulses in the plasma emission both in terms of the intensity of the emission lines and in terms of lifetime of the emission. Differences become significant only at very low fluences approaching the plasma formation threshold, which is significantly lower for 50 ps pulses than for 10 ns pulses. Calculations using a plasma ablation model show that initial plasma conditions are significantly different for 50 ps and 10 ns pulses during irradiation by the laser pulses. However, the dominant process leading to plasma emission at later times is from expansion and cooling of the plasma plume in the form of a blast wave in the ambient air which is primarily dependent on the energy deposited in the plasma and not on the pulse length [42].

1.5. Ablation mechanisms

The density of free electrons can grow exponentially by electron impact ionization, which involves collision of bound electrons with free electrons. If the free electron is accelerated such that its kinetic energy exceeds the ionization potential of the bound electron, ionization of the bound electron could occur, resulting into two free electrons. This process can repeat itself and lead to an avalanche event. Plasma with a critical density is formed when enough bound electrons are generated through ionization of atoms. The plasma forms within the laser pulse, and starts to absorb incident laser

light. Consequently, the laser light coupling mechanism into the target is significantly altered. For ultra-short laser pulses, the electrons are first driven to very high peak temperatures, while initially the lattice temperatures remain low. When the critical density level is reached, the plasma oscillation frequency is equal to the laser frequency and plasma with significant absorption character is formed. The electron-phonon scattering time scale is of the order of 100fs in most metals and semiconductors, while the phonon–phonon scattering time is on the order of 10ps. Consequently, for pico and femtosecond laser based heating of solids, initially the electrons are not in equilibrium with the lattice and subsequently electron-lattice heating raises lattice temperature. Recombination during the laser pulse is negligible because it requires longer time relative to the pulse duration. The energy density initially deposited within the lattice is responsible for the material surface damage through plasma expansion and material evaporation. The actual material damage and ablation usually occurs a few ps after the expiration of the laser pulse [35]. Prevalence of multiphoton excitation and absence of interaction of the laser pulse with ablated material appear to be the main characteristics of the femtosecond regime [43].

During its temporal evolution, the recombination character dominates the plasma expansion, and hence LTE hypothesis cannot be completely assumed. Energy transfer duration from the electrons to ions by coulomb collisions is longer than the laser pulse duration. Therefore, the conventional hydrodynamic motion does not occur during the femtosecond interaction time. There are two forces which are responsible for momentum transfer from the laser field and the energetic electrons to the ions in the absorption zone: one is due to the electric field of charge separation and another is the ponderomotive force. The charge separation occurs if the energy absorbed by the electrons exceeds the fermi energy (which is approximately a sum of the binding energy and work function) and escapes from the target. The electric field of charge separation pulls the ions out of the target. At the same time, the ponderomotive force of the laser field in the skin layer pushes electrons deeper into the target. Correspondingly it creates a mechanism for ion acceleration into the target.

The main contribution to femtosecond laser interaction with metals are from the processes of material expansion due to fast heating and very fast energy absorption leading to mechanical stress [44-46]. The superheating and material ablation of metals

caused by ultrashort-pulse lasers are simulated by a model based on the concept of phase explosion [47], which is a rapid boiling process due to the homogeneous nucleation in a superheated volume of liquid near its critical state. For PLD applications, the film composition is precisely same as that of the target because of the explosion like removal of the material at the high power densities [48, 49]. For dielectrics, the dominant channel for free electron generation is either impact or multiphoton ionization depending on the size of the band gap [50]. The ablation threshold for dielectric in the ultra short laser-matter interaction regime must be higher than that for the metals, assuming that all the atoms in the interaction zone are at least singly ionized.

Interaction mechanisms induced by ultrashort pulses, can be treated by different approaches depending on the material, such as the two temperature model [51], coulomb explosion [52-54] and non-thermal heating [55]. Laser–target interaction models using a hydrodynamic code include the absorption of laser radiation, the electronic heat conduction, the electron-phonon or electron–ion energy exchange, as well as a realistic equation of state.

1.6. Features of femtosecond laser-matter interaction

By comparing the femtosecond and nanosecond measurements, it appears that reduced amount of ions are measured in the femtosecond-laser-induced plasma. This is consistent with the fact that femtosecond laser pulses are too short in time to be able to interact with the plasma they have created. In the femtosecond interaction regime of expanding plasma, there are different population species with specific velocities. The electron number density and temperature of fs-laser induced plasma decreases faster than ns-laser induced plasmas due to different energy deposition mechanisms [56]. Because of its short pulse duration, the fs laser beam does not interact with the laser-induced plasma [57, 58]. Femtosecond pulses are of particular interest for ablation as the pulse duration is less than the typical thermalization characteristic time of a few picoseconds [59]. At constant laser irradiance, the target heating, melting and vaporization increase with laser pulse duration, and this applies also to the densities of vapor and background gas atoms and ions, and electrons in the plume, as well as to the plume expansion velocity and temperature, because the laser fluence rises with pulse duration. On the other hand, at fixed laser fluence, the target heating and evaporation rate increase for shorter laser

pulses, because of the rise in laser irradiance. The results show that the ablation thresholds are low in the femtosecond regime and plasma plume is strongly forward directed. The kinetic energy of the species evolved during laser ablation is close to 1 keV in the femtosecond regime and a few hundreds of eV in the case of nanosecond pulses [58, 60].

Femtosecond laser-induced plumes are found to be much smaller and weaker in intensity than those induced by nanosecond laser pulses [35]. The plume temperature and electron density during or shortly after the laser pulse become higher for shorter pulses at fixed laser fluence, because of higher laser irradiance. However, at a certain moment in time, sufficiently long after the laser pulse, it is observed that the total laser fluence, and not the pulse duration, determines the plume behavior, because the plume and plasma characteristics look very similar for different pulse durations, at constant laser fluence [41]. The temporal dynamics of the continuum and the spectral lines depend much more on the laser fluence than on the pulse duration. The decay time of the continuum emission due to the bremsstrahlung, is about three times higher with a nanosecond pulse than with a femtosecond one. With respect to ns-LIBS, the fs-laser induced spectra are characterized by smaller spectral continuum contribution, shorter duration and better resolution of the spectral lines [61, 62]. LIP illuminated by ultra-short, ultra-intense optical pulses is a rich non-linear medium for the study of parametric production of electromagnetic radiation and collective phenomena such as x-ray generation, terahertz radiation, amplification and guiding of optical pulses.

In subsequent chapters, details of the work carried out on LIP from some solid targets using ns laser pulses and simulation of plasma characteristics are described.

1.7. References

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