Chapter 7. Conclusions

In this thesis the mechanisms responsible for material removal under the ultrashort (sub-ps and ps) pulsed laser ablation of transparent materials have been investigated. It was argued that ablation takes place after efficient energy deposition into the material following strong coupling between the laser radiation and the electronic system, and energy redistribution to the lattice due to surface charging, electron-phonon coupling and defects strongly coupled to the lattice. The high efficiency of ultrashort pulsed laser ablation is reflected by the high fraction of the energy deposited in the kinetic motion of the removed mass [RFP98]. The initial mechanism of excitation (the primary laser interaction) is observed to be a fast build up of a free electronic population in the conduction band due to avalanche ionization seeded by multiphoton absorption during the laser pulse duration and subsequent heating of the electrons in the laser field with phonon assistance. The electron photoemission and the numerous collisions between the free electrons and lattice vibrations will lead to energy deposition into the lattice, with consequently either non-thermal or thermal removal of particles.

Fig. 7-1 illustrates the basic processes in dielectrics when irradiated with intense laser beams.

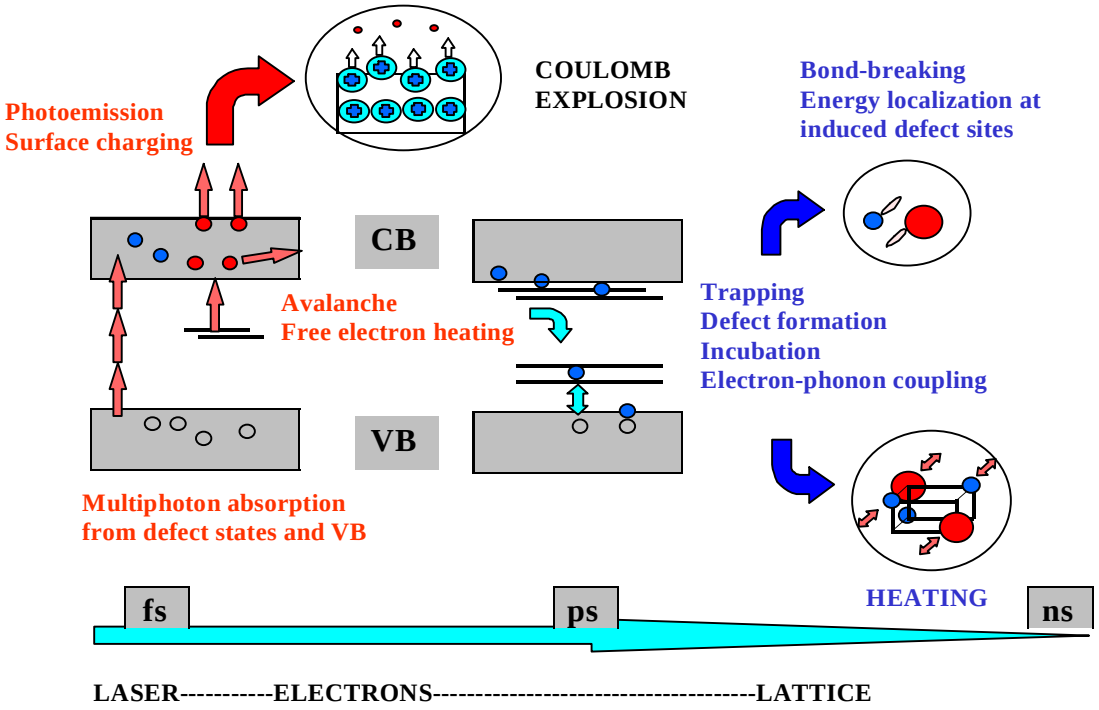

Fig. 7-1 Basic processes in dielectrics upon ultrashort pulsed laser excitation**.**

It has been shown that experiments with ultrashort pulse lasers (<5 ps) have considerable potential for studying the details of dynamical effects leading to particle removal from dielectrics since there is no complication due to the interaction of the laser pulse with emitted material. The potentiality of ultrashort pulses for fundamental studies has been illustrated by a large palette of investigations including ex-situ microscopy and in situ, time-of-flight investigations of the ablated species (ions, neutrals, electrons) emitted from transparent materials, mainly Al_2O_3.

Efforts have been made to identify the mechanisms responsible for material removal and the possibility of controlling them for practical purposes.

The experimental methods included static and dynamic experiments, for both identifying the processes and describing the time characteristics of the energy redistribution in the material.

We have indicated that macroscopic ion Coulomb explosion can occur from the surface region during the ”gentle phase” ablation due to high surface charging (average fractional charge of ca. 0.5) following electron emission, and subsequent electrostatic repulsion between the surface positive ions. TOF electron studies have indicated that the prompt electrons are responsible for triggering the Coulomb explosion mechanism. The time scale for this mechanism in Al_2O_3 is up to ca. 1 ps. The arguments for an electrostatic mechanism were extracted from measured values of momentum and energy for different species in the plume. As a signature for Coulomb explosion, chemical species with different masses in the same ionization states have equal momentum, and also the momenta scales with the charge for the same species in different ionization states, indicating that the impulsive acceleration mechanism is charge but not mass dependent. Coulomb explosion is restricted to a zone corresponding to the electron escape depth. No Coulomb explosion has been identified for semiconductor and metallic targets due to the fast charge neutralization in the electron depletion zone. The gentle phase is characterized by low ablation rates and high efficiency in ion production (more than 10% of the total material emitted), with high energies and narrow, forward peaked angular distributions. For laser pulse durations longer than a few ps, ion emission can still occur during the ”gentle” ablation phase but the distribution develops a dominant thermal contribution and the ions have a tendency towards the same kinetic energies rather than the same momenta. This slower ion emission is a consequence of electron-phonon coupling in the material leading to thermal excitation of the lattice (lattice heating) followed by a thermal particle removal.

It was indicated that the transition from the non-thermal gentle phase to the violent strong phase is closely related to incubation which results in a modified absorption cross section (and, hence, a decreased ablation threshold). After a certain accumulation period (related to the build-up of defects in the target material) the ablation behavior changes drastically. The irradiated surface becomes extremely rough, droplets and melting traces are found on the irradiated zone, and the maxima in the ion distributions move to lower velocities with a broadening of the angular distribution. During this ”strong” ablation phase, where there is evidence for a phase explosion mechanism, the ions have smaller velocities than the Coulomb explosion ions (observed for lower fluences and lower number of laser shots) and a tendency to equal energies. We interpret these ions as being a consequence of the decay of the gas phase ionized vapor plume (from which intense light is also emitted) after thermalization occurred. The strong phase exhibits high ablation rates and a low degree of ionization in the plume (ca. 1%). An overview of the ablation phases is given in Fig. 7-2. The appearance of a strong phase is a consequence of defect accumulation at multiple irradiation, which will enhance the probability for a violent removal of material of thermal nature due to the explosive boiling of the sample. Since dielectrics have low thermal transport properties, the conditions for achievements of high temperatures are met. The heating effect is very fast, on the picosecond time scale, due to efficient electron-phonon coupling and assistance of defects acting as trapping centers strongly coupled to the lattice.

It was also shown that, besides the increasing number of shots per site (with effect of incubation), also increasing the fluence or pulse duration moves the balance towards thermal effects, based on correlation with the electron times in the conduction band or, simply, with the ability to heat the system at high energy deposition rates.

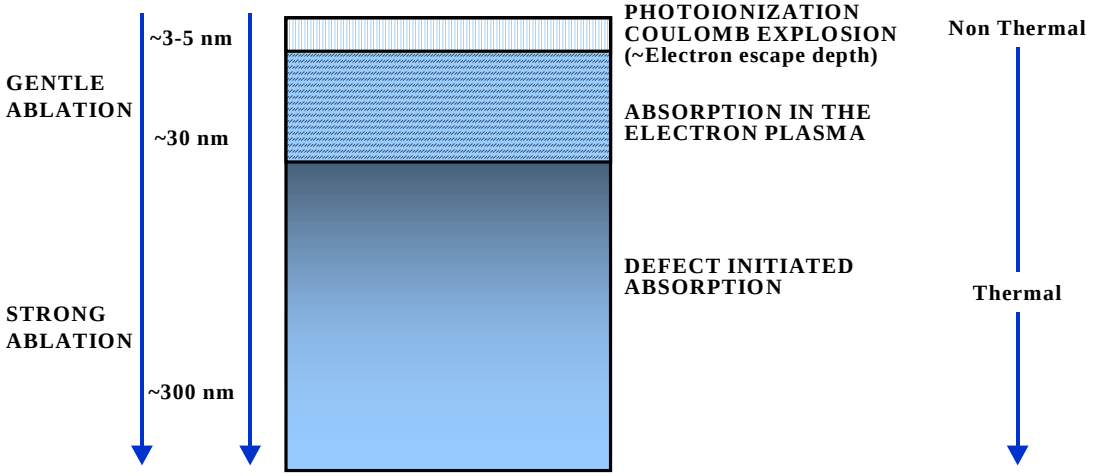

Fig. 7-2 Overview of the ablation phases at ultrashort pulsed laser ablation of dielectrics.

Pump-probe experiments allowed us to follow the time evolution of the energy deposition and redistribution process and the balance between electrostatic energy accumulation and heat deposition. Surface charging is found to survive beyond the critical value for surface break-up for about 1 ps. For the sapphire sample probe absorption is firstly maximized at about 600 fs due to better coupling with the hot electron system. Subsequently, at about 10 ps, the time scale of electron-phonon thermalization, the lattice appears to be most susceptible to absorption of the probe photons as a consequence of temperature increase. Finally, after several 100 ps the sample has cooled and loses the intermediate ability for very efficient energy absorption. For fused silica, heating takes place much faster in correlation with the strong electron-phonon coupling strength and electron trapping.

At the same time potential applications for microstructuring of dielectric materials have been presented in connection with the described processes. It has been show that the reduction in the unwanted thermal effects and the increasing of the localization in the energy deposition can lead to very clean structures induced in dielectric materials. The present studies open the way for controlled optimization of micromachining process in connection to the particular mechanisms for ejection (by employing suitable pulse shaping techniques and train pulses at THz repetition rate). Lattice heating by the first pulse and efficient absorption of a second pulse after some ten picoseconds might open new avenues to overcome the well known difficulties with deep hole drilling into transparent materials or surface polishing in precision optics fabrication. Cracks, which are observed even with femtosecond lasers, could possibly be avoided when ablation in the gentle phase regime is combined with energy deposition into the premolten material by a second pulse.