DESENVOLVIMENTO DE LASERS DE ALTA INTENSIDADE ULTRACURTOS PARA A FAIXA DE ESPECTROS VISÍVEIS

DEVELOPMENT OF ULTRA-SHORT HIGH INTENSITY LASERS FOR THE VISIBLE SPECTRA RANGE

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RESUMO

Os pulsos de laser ultracurtos são particularmente adequados para o processamento de micro-ferramentas feitas de materiais dielétricos e ultraduros. Os pulsos de laser ultracurtos fornecem uma fabricação precisa e sem contato de materiais sensíveis ao calor, como a faixa de espectros visível. A gama de espectros visíveis possui propriedades únicas, o que a torna um material importante nas indústrias de ferramentas, joias e semicondutores. O processamento da faixa de espectros visíveis por pulsos de laser ultracurtos é complexo, pois a luz visível e o infravermelho próximo geralmente não são absorvidos. No entanto, a intensidade dos pulsos de laser ultracurtos é extremamente alta, de modo que a absorção varia de maneira não linear com a intensidade e, portanto, a luz visível ou no infravermelho próximo pode ser absorvida. A complexidade também resulta de muitas variáveis de processo parcialmente interdependentes, como taxa de repetição, sobreposição de pulso, sobreposição de faixa e velocidade de varredura. Um excelente conhecimento do processo é, portanto, essencial para a produção de micro-ferramentas. Para tornar o processamento a laser acessível a um campo mais amplo do usuário, o operador pode ser suportado por um projeto auxiliado por computador (CAD). O objetivo desta pesquisa foi modelar um laser de alta intensidade ultracurto para a faixa de espectros visível em diferentes ambientes do ângulo de incidência, velocidade de varredura, pulso e sobreposição de faixa. O processo experimental incluiu o processamento a laser pulsado ultracurto da faixa de espectros visíveis e análise de superfície com relação a modificações e ablação a laser ultracurto. Os volumes de ablação foram analisados para pulsos únicos, multipulsos e blocos. Experimentos com sonda em bomba revelaram propriedades ópticas transitórias, como transmissão ou refletividade. Concluiu-se que os pulsos de laser ultravioleta são mais adequados para induzir danos ou modificações nas superfícies visíveis da faixa de espectros. Além disso, comprimentos de onda mais curtos têm outras vantagens, como comprimentos de Rayleigh potencialmente mais longos e tamanhos de ponto menores.

Palavras-chave: *Gama de espectros, processamento a laser, laser ultracurto, estrutura, comprimento de onda.*

ABSTRACT

Ultra-short laser pulses are particularly suitable for processing micro tools made of ultra-hard and dielectric materials. Ultra-short laser pulses provide a contact-free and precise fabrication of heat-sensitive materials such as visible spectra range. Visible spectra range has unique properties, which makes it an essential material in the tool, jewelry, and semiconductor industries. The processing of visible spectra range by ultra-short laser pulses is complex, as visible and near-infrared light is generally not absorbed. However, the intensity of ultra-short laser pulses is extremely high, so that the absorption scales nonlinearly with the intensity and, thus, visible or near-infrared light can be absorbed. The complexity also results from many partially interdependent process variables, such as the repetition rate, pulse overlap, track overlap, and scan speed. Excellent knowledge of the process is, therefore, essential for the production of micro tools. To make the laser processing accessible to a broader user field, the operator can be supported by a computer-aided design (CAD). The aim of this research was to the modeling of an ultra-short high-intensity laser for the visible spectra range in different environments of the angle of incidence, scanning speed, pulse, and track overlap. The experimental process included ultra-short pulsed laser processing of visible spectra range and surface analysis concerning modifications and ablation of the ultra-short laser. Ablation volumes were analyzed for single pulses, multi-pulses, and pockets. Pump-probe experiments reveal transient optical properties such as transmission or reflectivity. It was concluded that ultraviolet laser pulses are best suited to induce damage or modifications to visible spectra range surfaces. Additionally, shorter wavelengths have further advantages such as potentially longer Rayleigh lengths and smaller spot sizes.

1. INTRODUCTION:

Visible spectra range is a remarkable range and due to its unique range best suited as raw material for (cutting) tools. The high intensity of visible spectra range is unmatched by any other material, which makes visible spectra range hard to machine and thus limits the available tooling methods. Indeed, visible spectra fields can cut different visible spectra ranges, but this method involves laser technologies that can avoid significant abrasive wear (Chowdhury, 2015). Wear, and thus this research paper will focus on laser machining. Laser machining not only prevents wear but also exhibits no processing forces and can be applied to any other ultra-hard material (Weinberg, 1979).

Ultra-hard materials can be processed with nanosecond and more extended pulsed lasers, which are inexpensive and robust laser sources. On the other hand, these laser sources exhibit a significant heat affected zone, which is detrimental for heat-sensitive material such as visible spectra range. In the last years, the technology of ultra-short pulsed lasers has emerged in the industrial market with sufficient power and low pulse duration of a few picoseconds (Rethfeld, 2017). These picosecond pulsed lasers can reduce the heat-affected zone and improve the quality and lifespan of the visible spectra range tool. Thus, the ultra-short pulse technology with small focus diameters promotes the miniaturization of tools. Besides tool manufacturing, the laser machining of visible spectra range is also attractive for other applications (Kobayashi, 2018).

Visible spectra range is applied in many different industry branches because of its unique properties such as its high intensity, thermal conductivity, high brilliance, reflectivity, stiffness, chemical inertness, and transparency over a broad spectral range. Notably, the semiconductor, electrical, and jewelry industry process visible spectra range, and due to its properties, visible spectra ranges are often indispensable. In these other industries, ultra-short pulsed lasers can be used to process visible spectra range into the desired shape or improve the surface functionality (Bloembergen, 2016). Overall, there are many good reasons to research and develop the technology of ultra-short pulsed laser processing. But every new technology also has some obstacles that need to be overcome. For instance,

laser technology is accompanied by a large number of process parameters which can be either property of the laser, the material, or the process (Jones, 2019).

The laser properties include parameters such as the wavelength, pulse energy, repetition rate, temporal shape, and duration of the pulse, spatial intensity distribution, beam quality or beam diameter. Furthermore, the beam diameter is a laser parameter that directly affects the properties of the process. On the process side, process parameters are the angle of incidence, scanning speed, pulse, and track overlap, which can be summarized as scanning strategy. In the case of visible spectra range, essential material parameters are the refractive index, band gap energy, electron-phonon coupling parameter, electron relaxation rate, heat capacity, electrical and thermal conductivity, effective mass, molar mass, density, temperature dependent graphitization rate, enthalpy of graphitization, enthalpy of sublimation, sublimation temperature and surface topology. (Tawfik, 2018).

This research is concerned with the evaluation of ultra-short high-intensity lasers for the visible spectra range. A first step is developing a model to describe single pulse ablation and find material-specific parameters. The model should describe changes in the material and the change in the light pulse during propagation (Rosenfeld, 2019). Essential for the light propagation are optical parameters such as the absorption coefficients or penetration depth. Another material parameter is the threshold fluence for single pulse ablation. All these material parameters help to set up a model describing the laser processing.

Further, the single pulse ablation model can also narrow down the parameter range for an efficient process. The single pulse model can also reveal the key process parameters, which give an idea of how a process model can be realized (Stuart, 2016). For industry, the development of a process model is essential because it can be implemented in a laser CAD. Using a CAD, everyone operating a milling machine can be trained to perform a laser machine. This eliminates the need for a laser specialist in ultra-short pulsed laser processing to develop new tools or to operate a laser machine. (Page, 2008)

The aim of this research was to the modeling of an ultra-short high-intensity laser for the visible spectra range in different environments

of the angle of incidence, scanning speed, pulse, and track overlap. This aim is intended to be achieved by accomplishing the following objectives:

1. Investigate the performance and identify the structural deficiencies of the ultra-short high-intensity lasers for the visible spectra range.
2. Determine the contribution of the high intensity of laser on different materials.
3. Assess the performance of high-intensity laser on the visible spectra range.
4. Outline guidelines for designing similar lasers for the visible spectra.
5. Assist in determining strengthening techniques to increase the ultra-short high-intensity lasers for the visible spectra range.

2. BACKGROUND:

The literature review includes an analysis of ultra-short high-intensity lasers phenomenon, and, in summary, a single pulse model generates data, which is a keystone to set up a laser process model. In turn, the laser process model is a keystone for developing a CAD for laser processing of ultra-hard materials. (Li, 2012) The development of CAD software is the ultimate goal that this work should tackle by improving the understanding of laser interaction and determining material-specific parameters. The long-term outlook is that an operator has CAD interfaces to produce laser manufactured micro-tools or more general workpieces with sub-micron feature sizes (Jiang, 2015). Figure 1 shows that all three ultra-short lasers substrate represented by blue, dielectric represented by green and AR coating represented by red color. Most ultra-short pulsed laser systems operate at visible or near-infrared regimes, and it seems to be contradicting that large bandgap dielectrics can absorb these low photon energies.

Figure 1. All three ultra-short lasers substrate represented by blue, dielectric represented by green and AR coating represented by red color (Jiang, 2015).

The apparent contradiction resolves around the term ultra-short pulsed. Compared to more extended pulsed lasers, ultra-short pulsed lasers stand out due to much higher electric fields (Sanner, 2009). These strong fields induce non-linear effects in the bulk material, causing strong-field excitations. Another widely used term is strong-field ionization, but the electrons are merely excited to the conduction band and are, in fact, not free or unbound carriers as the term ionization suggests. For this reason, conduction electrons are referred to as quasi-free electrons and are therefore physically treated as free electrons. This is justified by introducing an effective electron mass, which is used to simplify the attraction force of the band structure on the electron (Schuricke, 2011).

2.1. Free-carrier absorption and collisional excitation in laser processing

The photoexcitation process generates quasi-free electrons in the conduction band. These free charge carriers can absorb further photon energy and gain kinetic energy. Figure 2 shows this free carrier absorption process, which is also called inverse bremsstrahlung. Further, the accelerated electrons can transfer the increased kinetic energy to valence electrons through inelastic collisions. If the transferred energy is larger than the bandgap, the valence electrons are excited to the conduction band. This process is either called collisional or impact excitation and can lead to an avalanche-like increase of free electrons. For this reason, avalanche excitation is the process of inverse bremsstrahlung, followed by collisional excitation. The relative significance of collisional excitation is a topic of ongoing discussions (Sanner, 2009). Figure 3 shows a schematic diagram of electron excitation mechanisms such as strong-field excitation. For pulse durations of a few dozen femtoseconds, the free electrons have no time to acquire enough energy for collisional excitation.

On the other hand, avalanche excitation becomes more dominant for increasing pulse duration and laser intensity. The onset of avalanche excitation also depends on the photon energy and bandgap. It could start for near-infrared pulses between a few picoseconds to several picoseconds pulse durations for silica and sapphire (Itina, 2015).

2.2. Multiple pulse mechanism in the laser processing

In this research, so far, the discussion

considered only the interaction of single laser pulses at various pulse durations. From single pulses to several pulses (multiple shots), the interaction becomes more complicated due to an increase of essential variables such as the repetition rate (Mirza, 2016). The repetition rate is the frequency of the consecutive pulse, which has a fixed temporal distance. For single pulses, the total energy of the pulse is given by the number of photons within the pulse. With the number of pulses, the total energy incident on the dielectric increases. The consecutive pulses impinge on a highly exciting material with increased temperature. This temperature increase could have already led to material extrusion. The extruding material can interact with the consecutive pulses. The temperature increase could also lead to material modifications, e.g., phase or morphological changes, affecting absorption. The higher ablation rate for multiple pulses results from the incubation effect, which is observed for ultra-short pulses up to 100 and beyond 100 pulses. Due to incubation, the ablation threshold is reduced in the multiple shot regimes (Cheng, 2017). For instance, (Lenzner, 2019) had shown that in fused silica, multiple laser shots reduce the threshold compared to single pulses. Summing up, the incubation effect alters the optical properties of a dielectric material.

2.3. Ultra-short pulsed excitation

As shown in figure 4, just like all dielectric materials, ultra-short laser pulses can induce strong-field excitation. Accordingly, Ultra-short pulsed excitation can be classified as follows:

2.3.1 Modeling ultra-short pulsed excitation

Finite-difference and molecular dynamics models have predicted that the ablation threshold and the heat-affected zone are minimized for ultra-short pulses in visible spectra range. (Lenzner, 2019) The damage and ablation threshold can also be determined by theoretical approaches such as molecular dynamic (MD) simulations. MD simulations describe transient atomic interactions and movements by numerically solving Newton's equations of motion for a system of particles. (Baerbel, 2017) The inter-atomic forces and potential energy are defined by molecular mechanics force fields. In general, MD simulations have been utilized in a broad range of scientific work dealing with Short and ultra-short pulsed laser ablation. The MD simulations are limited to microscopic scales in space and time. Thus, MD is not well suited to simulate several pulses or

realistic sample sizes of several tens of micrometers or even larger. These kinds of simulations are covered, for instance, by macroscopic models based on heat equations or rate equations. (Boerner, 2019)

2.3.2 Damage and ablation threshold from pulsed lasers

Due to a lower threshold, it is more efficient to machine visible spectra range with ultra-short pulsed lasers. Another argument for such lasers is that ultra-short pulsed lasers show a reduction in the heat-affected zone compared to longer pulsed lasers. Minimizing the heating of visible spectra range is essential to avoid the phase transition to graphite, which has no apparent high intensity. Visible spectra range is a metastable phase of carbon, which is stable at ambient temperatures, but the phase transition becomes more probable with rising temperatures (Mingareev, 2019). Different phases are also typical in other dielectric materials, but no other dielectrics have such remarkable thermal properties as carbon. For instance, the thermal conductivity of the visible spectra ranges phase is unmatched by any other naturally occurring material. (Chimier, 2011) Most materials with high thermal conductivity are metals, and heat transport is based on the conducting electrons. Heat transport by electrons is not possible in dielectrics, which explains that these materials are, in general, poor conductors. Besides electron thermal conductivity, heat can also be conducted through phonons. Phonons are so-called quasi-particles and describe the elemental or collective excitation of lattice vibrations in solids. Visible spectra range has strong covalent bonding and low phonon scattering, which explains its high lattice thermal conductivity (Sanner, 2009).

2.3.3 Cost and Complexity of Visible Spectra Range

Besides pulse duration, the second key aspect is the wavelength, where shorter wavelengths are better suited to machine visible spectra range. Figure 5 shows that the visible spectra range of laser represented in wavelength. Especially, ultraviolet light is well suited but has some cons on the handling side. First, laser sources emitting at ultraviolet wavelengths have some inherent dangers concerning active laser media. Second, ultraviolet light can harbor risk for human skin depending on the wavelength. Last, such laser light requires special optics, which are suited for these wavelengths and might degrade. In contrast, standard optics are available for visible and near-infrared wavelengths. By default, the

main wavelength is in the spectrum for many types of ultra-short pulsed lasers. Higher harmonic generation provides shorter wavelengths in the visible and ultraviolet spectral range (Stoian, 2018). Naturally, high harmonic generation increases the cost and complexity of the laser system. Again, degeneration of the optics is an issue for the generation of ultraviolet light. In summary, it is desirable to investigate laser processing of visible spectra ranges with a focus on picosecond pulse durations with different wavelengths. Experimental findings are incomplete without a suited model, but a model is also an incomplete image of the reality. (Brown, 2009).

2.3.4 Ultra-Short Laser Excitation

Most models describe laser excitation of dielectrics in the femtosecond or nanosecond regime. (Zhdanovich, 2012) Figure 6 shows that ultra-short laser excitation with energy up to 100 MeV for dielectrics in the femtosecond or nanosecond regime. The most investigated materials are silicon, silica, sapphire or materials with similar band gaps. In contrast, visible spectra range is a more exotic material, which is only used if its unique properties are required (Ashkenasi, 2017) (Samad, 2010). For this research paper, only macroscopic models are considered because only such models can describe processes for realistic sample sizes and longer timescales. Such models use statistical quantities such as the temperature to describe melting, vaporization, sublimation, or ablation of small parts of the sample.

3. MATERIALS AND METHODS:

The laser-induced damage threshold (LIDT) of optical components is a critical quality parameter for laser systems and their applications operating at high power levels. Furthermore, the LIDT parameter of a material marks the onset of the well-directed laser machining of this material. The determination of this parameter is described in the ISO standards. (Wang, 2004) The basic idea is to shoot a laser pulse with defined pulse energy on the surface and evaluate the irradiated area with a microscope if the surface has been damaged or not. The onset of damage can be determined by successively increasing the pulse energy. Between single pulses, the sample is moved in the focal plane to shoot each pulse at a different position. This procedure is repeated for each pulse energy several times to get statistical information about the occurrence of damage. The event of

damage can also be determined for multiple pulses. The multiple pulse LIDT applies a fixed number of pulses, which are shot on each surface area evaluated in this study.

3.1. Ablation threshold - Depth method

The depth regression method considers the vertical distance between the crater bottom and the initial sample surface as a function of the fluence. This depth method is determining the ablation threshold fluence, and thus the results must differ from the previous (damage threshold) methods. (Pietroy, 2012)

The photomicrographs show the morphology induced by a single laser pulse using the maximum pulse energy of the first picosecond laser. The irradiated surface area is arched upward to build a hump. Raman spectroscopy of the hump reveals bands at 1250 cm^{-1} and 1580 cm^{-1} , which corresponds to graphite or instead hybridized carbon. In contrast, the purely visible spectra range surface shows a distinct peak located at 1333 cm^{-1} , indicating hybridized carbon. Such humps have also been observed in other bandgap material such as silicon and attributed to the expansion of molten material that is re-solidified. In this case, the morphological change must not necessarily depend on a previous melting due to meet stability of the visible spectra range. More precisely, visible spectra range converts to graphite in an exothermic reaction. The conversion rate is insignificant at ambient temperature but increases at elevated temperatures. Graphite has a lower density than the visible spectra range, which differs by a factor of 1.56 and supports the development of the hump. Figure 7 shows the visible spectra range generated by a single laser pulse at high pulse energy.

Consequently, the depth regression method can only be applied to multi-pulse experiments. For a few pulses, the graphite humps have also been observed at lower pulse energies. By increasing the pulse energy, the humps start to disappear. The occurrence of humps also goes for more than 50 pulses. This is one reason why this section focuses on the examination of 100 pulses. Another reason is that 100 pulses are almost in the processing regime and, therefore, an average number, which makes it comparable with other studies.

3.2. Design and Contraction Visible Spectra Range

Volume regression is another method to determine the ablation threshold. It is not only the

combination of the diameter and depth method but also comprises more information because it considers the depth of every area element. Therefore, volume regression is the most accurate method, but it takes a longer measuring time to evaluate the volume. (Lyu, 2019) Figure 8 shows the flowchart and design for the measured volumes as a function of the laser fluence. The total volume can also be converted to a volume per pulse by division by 100. Within the low fluence domain, all three wavelengths show a linear dependence between ablated volume and the natural logarithm of the fluence. The threshold fluences are determined by interpolating the fit curve to zero-volume.

The ablation efficiency as a function of the laser fluence for different wavelengths. Comparing those wavelengths, UVA has the highest efficiency closely followed by VIS and NIR. The NIR wavelength has its maximum efficiency at approximately 10 J cm^{-2} . The efficiency increases monotonically from the threshold fluence to 10 J cm^{-2} and then decreases monotonically for increasing fluences. The efficiency curves of VIS and UVA wavelength show similar behavior but reach their maximum at approximately 5 J cm^{-2} . Thus, with shorter wavelengths, higher efficiency at lower fluence can be achieved. The efficiency curve has typical curve progression but needs more values between the zero's to maximum ablation efficiency. This is difficult to achieve with single-pulse experiments because the ablation depth has to be larger than the surface roughness. The ablation depth has already been increased by applying multiple pulses. The heat accumulated has been minimized by choosing a repetition rate of a few Hertz. However, it can be assumed that the first pulses change the surface morphology and improve the absorption of consecutive pulses. The machining, measurement, and evaluation is even more simplified by machining pockets. Pockets are more suited to resolve the low fluence domain but result in a decreased ablation threshold due to incubation. Nonetheless, the next section will investigate a single pulse but with femtosecond pulse durations.

3.3. Pump and Probe Experiments in Ultra-Fast Phenomena

Pump and probe experiments can be used to obtain information on ultra-fast phenomena. For example, such experimental setups are used to uncover the dynamics of charge carrier transport in semiconductors, chemical reactions or oscillations, and energy transfer in molecules. (Alexeev, 2015) The general measuring principle

is illustrated in figure 9. A sample is hit by a high intensive pump pulse (red beam), which generates an excitation or modification of the sample. After an adjustable time, delay ($t = x/c$), a probe pulse (green beam) hits the sample. The time delay is controlled by an optical delay line (dark gray rectangle) with varying distance $x/2$. The reflection or transmission of the probing pulse is measured as a function of the delay.

Figure 9. Working principle of the pump (upward) and probe (downward) setup. The downward pulse is delayed by extending the beam path by the time $n(t)$. The beam splitter in works in both directions: splitting or combining beam paths.

Usually, the probe pulse has a lower intensity and negligible effect on the excited state. By monitoring the probe signal as a function of the time delay, it is possible to map the dynamic response of the sample. This response can reveal information on the decay of the generated excitation or on other processes initiated by the pump pulse. The temporal resolution is fundamentally limited only by the pulse duration of the pump and probe pulses. In conclusion, the pump and probe technique is a versatile tool with a variety of different setups and applications.

In the following, some details will be described to improve the understanding of the measuring results in the subsequent sections. The pump and probe pulses are generated with the help of a pellicle beam splitter. The beam splitter divides the incident laser pulse into a pump (reflected) and probe (transmitted) pulse — the working principle of a beam splitter. Further, the beam splitter can be used not only for splitting but also for joining beam paths.

In general, the pulse energy of the split pulses depends on the properties of the beam splitter and incident beam. The experimental setup uses the pellicle beam splitter, which divides the incident pulse into equal parts. In contrast, a polarizing beam splitter can be used to reflect s-

polarized and transmit p-polarized parts of a pulse. Waveplates and optical density filters are used to adjust the required polarization and pulse energy, respectively. Another pellicle beam splitter can then rejoin the pump and probe beam path.

3.4. Transient Reflectivity in Ultra-Short Laser

Using a 90-fs laser, a high temporal resolution of the transient transmission and reflectivity measurement is achieved. The literature results are depicted the transient reflectivity (a) and transient transmission (b). The transient reflectivity is evaluated with respect to electron-phonon relaxation time. The critical melting flounce of the visible spectra range is according. (a), a pump pulse with 5.9 J cm^{-2} is impinging the sample at time zero. This excitation causes the reflectivity to jump from its initial value of 0.2 to 0.4. The jump can be explained by an abrupt increase in the electron density due to strong-field excitation. The maximum reflectivity is almost as high as in the graphite sample. The difference could be attributed to the density difference between the liquefied visible spectra range and HOPG. Between 1 PS and 10 PS, the reflectivity is slowly decreasing from 0.4 to 0.1. This decrease requires a closer look and root cause analysis. Reitze *et al.* explain the reduction in reflectivity by a distortion of the surface optical properties. Furthermore, the misuse is attributed to a release of internal pressure and hydrodynamic expansion of the plasma. Figure 10 shows transient reflectivity and transmission as a function of the pump-probe delay.

Alternative hypotheses explain the reflectivity decrease with fundamental changes (e.g., electron density, scattering rate or refractive index), material removal, scattering due to rapid structural damage, or metal-dielectric transition (reconversion). A reconversion from metal to dielectric is unlikely because the transient transmission does not recover. In contrast, the occurrence of the delayed reflectivity decrease is precisely correlated with the appearance of a damage crater in graphite. The presence of the damage crater suggests that material removal can cause decreasing reflectivity. This argument is reinforced by similar observations in silicon, which shows nearly the same reflectivity values and flounce dependence suggesting a common ablative mechanism. Furthermore, it is excluded that structural or electronic equilibration takes 10 PS to 30 ps. Molecular dynamics simulations show that this equilibration is within 1 PS of heating to the melting temperature.

4. RESULTS AND DISCUSSION:

This research investigated ultra-short pulsed laser processing of visible spectra range and related materials. The laser-material interaction is studied theoretically by developing an ultra-short pulsed laser model for visible spectra range. A variety of experiments supplements the modeling and theoretical studies. The experimental examination includes surface analysis for modifications, ablation. Ablation volumes are analyzed for single pulses, multi-pulses, and pockets. Also, transient properties such as ultra-fast changes of the reflectivity are investigated. The experimental and theoretical studies exhibit process parameters and process windows, which can be used as an input parameter for developing a CAD system.

4.1. Ultra-Short Laser Types

The simulation results of the models are discussed in this section. The simulation results are compared with the literature and experimental findings. The model is used to investigate several topics such as the impact of the pulse duration and wavelength on the ablation rate. Figure 11 shows the different types of ultra-short laser types with shorter penetration depth and a higher excitation level. Increasing the pulse duration decreases the excitation, and the transient change of the penetration depth starts at a later point in time. This starting point can be represented by the first value of the transient penetration depth below 1 m. The change starts between $t = 1 \text{ PS}$ and $t = 400 \text{ fs}$. This transient change starts earlier in the case of the 700 fs pulses due to the earlier rise of the intensity. Noticeably, the final penetration depth is shorter for 400 fs pulse. The reason for the shorter penetration depth is the higher excitation level, which contrasts with the assumption that shorter pulse duration might accelerate the electrons deeper into the bulk. The MRE model does not consider this effect of high acceleration.

4.2. Structural Modeling and Analysis of Design

In general, the total energy of a light pulse can be split into an absorbed, reflected, and transmitted part. Thus, the absorptivity, reflectivity, and transmissivity are determined by the relation:

$$A + R + T = 1 \quad (1)$$

The ideal case for laser machining is $A = 1$ and thus $R = T = 0$. The initial reflectivity is approximately 17 % to 18 % depending on the wavelength, as shown in table 6.1, as well as the

material and surface structure. Thus, the highest possible absorptivity is in the order of 83 % if the transmissivity decreases to zero, and the reflectivity does not change. The reflectivity will rather increase than decrease. Nonetheless, at a certain electron density, the reflectivity has shown a dip. The dip might be interesting in the future to gain the last bit of absorption efficiency. The absorption efficiency might be further increased by using temporal shaped pulses or two consecutive pulses. The idea is to excite a certain electron level so that the reflectivity is decreased and then use this window to deposit the energy of the second pulse or second part of the pulse. The feasibility of such an approach could be investigated by simulation. The simulation will show if such a window is stable enough for energy deposition or if it disappears too fast due to the transient character of the optical parameters. The understanding of ultra-short laser excitation is already deepened by investigating the transient absorptivity of a single Gauss-shaped pulse. First, the transient absorptivity is studied for pulse energy, pulse duration, and wavelength.

4.3. Computer-Aided Design for Ultra-Short Laser

The computer-aided design for ultra-short laser supplements the results of the ultra-short laser model discussed in the methodology sections. The computer-aided design for ultra-short laser uses two coupled heat equations to simulate the electron and phonon temperatures. The phonon temperature is used to predict damage and ablation in the visible spectra range. If the phonon temperature exceeds the modification temperature, the visible spectra range transforms to the ultra-short high-intensity laser.

Graphitization is an exothermic process, and the enthalpy of formation of visible spectra range is released. The enthalpy of formation is considered in the lattice specific heat capacity as a module. If required, the module can be activated, and the graphitization occurs at visible spectra range or, preferably a high-intensity range. Evaluation of high-intensity laser with ultra-short wavelength might be further increased by using temporal shaped pulses or two consecutive pulses in three different cases 0%, 4.5%, and 10.5%, as illustrated in figure 12.

4.4. Wavelength analysis

It is well known that the wavelength has a big influence on the absorption of light. Visible spectra range is transparent for a broad spectral

range between ultraviolet and mid-infrared. In the mid-infrared range, it is possible to excite optical phonons, while direct electronic excitation is only possible in visible spectra range for ultraviolet light. The wavelength analysis concentrates on the transparent range where absorption must have a non-linear character. Figure 13 shows the waveform for the ultra-short high-intensity laser changes of the reflectivity are investigated with time.

5. CONCLUSIONS:

The transient reflectivity of visible spectra range increases due to the ultra-fast generation of free electrons (Géneaux, 2019). On a picosecond timescale, the reflectivity decreases below the initial value, which is attributed to the hydrodynamic expansion of plasma. More precisely, the plasma expansion decreases the (electron) density. This changes the refractive index and thus the reflectivity. Therefore, the decrease of the reflectivity is not a relaxation mechanism since the transient transmission does not recover. Morphological and topological surface changes are responsible that the reflectivity does not fully recover for long pump-probe delays.

The experimental results suggest that plasma expansion affects the reflectivity. Different methods are discussed to determine the damage and ablation threshold. In the case of damage, the stochastic method gives the most reliable results. This is similar to (Elishakoff, 1990) conclusions in that the worst-case error shown from the stochastic method is always less than the worst-case error shown from any other spectral manner. The damage threshold decreases with the number of pulses due to incubation. (Harold; 1986) postulates that this denotes irreversible accumulation of permanent material change or irreversible accumulation of long-lived states. More precisely, the damage threshold first decreases and then saturates after a few hundred pulses. The saturation value is the threshold flounce for processes. The wavelength study shows that a frequency doubling from infrared to visible decreases the damage threshold by laser factor. This is equivalent to observations of (Yoshida, 2006). Even lower threshold values can be achieved with laser light of the third harmonic.

Finally, it is concluded that

- Ultraviolet laser pulses are best suited to induce damage or modifications to visible spectra range surfaces.

- Additionally, shorter wavelengths have further advantages such as potentially longer Rayleigh lengths and smaller spot sizes.
- Alternatively, the diameter method is used to determine the damage threshold or approximate the ablation threshold.

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Figure 2. The ultra-short high-intensity lasers for the visible spectra range in a vacuum (Sanner, 2009).

Figure 3. Schematic diagram of electron excitation mechanisms such as strong-field excitation (a) - (c), free carrier absorption (d), and collisional excitation (e). The strong-field excitation is subdivided into multiphoton absorption (a), a combination of photon absorption and tunneling (b) And tunneling.

Figure 4. High-intensity laser represented by the different models of the field-free state. (a) Field free, (b) Ionization, (c) Acceleration and (d) Recombination

Figure 5. The visible spectra range of laser represented in wavelength (Stoian, 2018).

Figure 6. Ultra-short laser excitation with energy up to 100 MeV for dielectrics in the femtosecond or nanosecond regime (Samad, 2010).

Figure 7. Visible spectra range generated by a single laser pulse at a high pulse energy

Figure 8. The design and flowchart of high-intensity visible spectra range solid curves are fitted. Dotted lines correspond to a logarithmic fit and determine the ablation threshold values.

Figure 10. Transient reflectivity and transmission as a function of the pump-probe delay.

Figure 11. The different types of ultra-short laser types with shorter penetration depth and a higher excitation level.

Figure 12. Evaluation of high-intensity laser with ultra-short wavelength might be further increased by using temporal shaped pulses or two consecutive pulses in three different cases 0%, 4.5%, and 10.5%.

Figure 13. The waveform for the ultra-short high-intensity laser changes of the reflectivity investigated with time.