Investigation of the Material Removal Efficiency During Femtosecond Laser Machining

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Abstract

Understanding physical processes accompanying ablation is necessary for the optimal use of Femtosecond Laser pulse material processing. The current work describes the implementation of a molecular dynamics based approach, developed for simulating the Laser ablation process of iron at a molecular level. A measure of material removal is formulated and utilized to estimate the effect of energy and pulse width on material removal. Evaluation of the model is based on comparison with experimental results. Ablation efficiency dependence of Laser Fluence is discussed.

Keywords: Laser ablation efficiency; Molecular Dynamics; Femtosecond laser

1. Introduction

Laser micro machining is a technology capable of producing parts in the micro and sub micro scale. For such applications, lasers with pulse duration in the femtosecond range (femtolasers) are widely used. Laser ablation occurs due to the irradiation of the laser beam onto the material causing a combination of sublimation, vaporization and melting. It is characterized by small temporal and spatial scales and an extremely high material temperature and pressure. Femtolaser (FL) drilling uses pulses with duration in the order of tens to hundreds of femtoseconds. During the recent years, the FL ablation has shown a rapid development due to its distinct advantages compared with those of the longer pulse lasers [1]. As a result of its extremely short pulse duration, the heat diffusion is confined, and the heat affected zone (HAZ) is rather limited. This high localized heating, in each laser pulse, results in smaller material volume removal and therefore, more precise machining results compared with the ones obtained from longer laser pulses. The laser ablation process depends on the physical properties of solid matter, laser parameters and environmental conditions [2]. Experimental investigations have revealed that with FL, along with the crater, co-exists a debris formation, the height of which depends on the target matter, the laser energy and the number of pulses fired to the target. The crater diameters have been found to be greater than those of the laser beam. Coincidence on the crater shape with radiation intensity distribution has been found only at low laser fluences ($\approx 1 \text{ J/cm}^2$). The crater widening is the result of the laser beam being scattered in the near-surface plasma and the plasma action on the crater formation. The debris generation is the result not only of the liquid phase explosion, but also of the plasma particle recondensation [3]. Laser micro-processing is used by many industrial sectors – semiconductors, electronics, medical, automotive, aerospace, instrumentation, and communications. Laser ablation of metals, ceramics, silicon and polymers is a complex process and the exact nature of their interaction with the material and laser processing parameters used, is specific.

FL being a rather precise tool needs accurate simulation methods [4]. Molecular Dynamics (MD) is based on the solution of Newton’s second law and aims...
at monitoring the movement of an atom within a system. Each particle in the ensemble of N particles is treated as a mass point, interacting through force fields, which in turn, have been derived from interacting potentials. MD works through the solution of Newton’s equation of motion. Therefore, the evolution of the system can be calculated in certain time steps, where the total information (particle positions, velocities, kinetic and potential energies) of the system is available for each time step. All further properties as temperature, pressure and autocorrelation functions can be determined without any additional parameters [5]. The reaction process of a laser beam with a bulk solid is the interaction between the electromagnetic field of light and the electric field of the atoms or molecules of the solid material. The light absorbed by the solid material can be divided into three categories depending on the level of the light energy: 1) The excitation of the electronic state of atoms and molecules of the lattice particles, 2) The excitation of the intermolecular vibration state of the lattice molecules and 3) The excitation of the intermolecular or interatomic vibration of rotational state of the lattice molecules.

The majority of the interacting potentials used in MD, are based on the Pair Potential Approximation (PPA). This is so, due to the fact that PPA provides a good description of the material’s properties and also consumes less computing time than do the potentials utilizing three or more body terms [7]. Potential functions that have been “phenomenologically” derived, often present a more realistic view of the atomic interactions than do the potentials having been exclusively derived. The following types of potentials fall into the phenomenological category: Buckingham, Morse, Lennard-Jones (Mie’s reduced form) and Barker potentials for Krypton and Xenon. Among the above potential functions, the Morse Potential Function (MPF) is considered to be more appropriate for its application to metals [5]. The embedded-Atom Method (EAM) is a semi-empirical, many-particle potential for computing the total energy of a metallic system. The influence [8] of the MPF and EAM based potential, on the description of the properties and the ultrashort laser ablation process of Fe by MD Simulation technique, has been studied. The accuracy of both potentials in evaluating the melting temperature, the linear thermal expansion coefficient and the compression behaviour of Fe, is calculated [9].

Numerous studies have been conducted for the decomposition of metals during the FL ablation, following different approaches, varying from analytical mathematical modeling to simulation and experimental studies. Studies have applied Hybrid MD Simulations, Classical MD or MD, combined with the Two Temperature Model (TTM), in order to investigate the laser ablation phenomena, such as the heat transfer mechanism and shock wave propagation [10], their dependence on the initial temperature, the pulse duration [11] and the mechanism and velocity distribution of molecules or plume, ejected during laser ablation [12]. According to [13], the results from the MD modelling of the ultrashort laser pulse ablation of metals (Al, Ni and Fe), are reported at fluences, and range from the threshold of ablation up to 0.5 J/cm².

In the present study we define as laser ablation efficiency the ratio of the ablated material volume to the energy of the laser pulse.

Section 2 provides the molecular dynamics model describing the implemented algorithm and the simulation details of model including the material and laser beam properties. In section 3 the experimental setup, that validates the model, is determined. The results from the molecular dynamics model and the experiments are presented and discussion in section 4 with special emphasis given on the laser ablation efficiency.

2. Molecular Dynamics

2.1. General Description

After energy has been absorbed by the material, the electro and lattice temperatures rise greatly. When the material temperature is high enough, certain phase change processes occur and part of the material is removed from the bulk. Complicated thermal and mechanical processes take place and many factors contribute to the material being removed. Temperature is the dominant factor that leads to material separation and consequently, to its removal. The electrons and the lattice are heated strongly by the laser irradiation in order for the increase in the lattice temperature to be high enough for melting and evaporation to occur. Since the lasers, used in micro/nano processing are operating in the femtosecond range, the irradiated material is assumed to be going from a solid to a gas phase without having first gone through a melting phase [19]. The target to be irradiated is considered to be of BCC structure, at an initial constant environment temperature. The lattice structure of pure Fe changes with the increase of temperature. Until 1118 K the crystal structure is off BCC. After this temperature has been reached and up to 1663 K the structure is transposed to FCC. From 1663 K and above the form is of BCC again. It is safe to assume that the crystal structure of pure Fe is constant in the BCC form, due to the ultrafast heating of the bulk material, provided by the femtosecond Laser pulses. The incident laser beam irradiates the target as shown in Figure 1, with a uniform temporal distribution. The laser beam propagates along the Z axis, perpendicularly to the surface of the target. The laser beam intensity is considered being spatially uniform and has Gaussian
temporal distribution. The number of photons, corresponding to the laser energy, is deposited to the material exponentially, following the Beer-Lambert law. This photon energy is transferred to the atoms of the system within a characteristic time, the time for the electron lattice energy transfer and the establishment of equilibrium temperature. The energy deposited to the system’s atoms contributes to their kinetic energy being increased. This work uses an MD developed approach implemented in a code, based on the Morse Potential Function for the simulation of laser ablation processes. The approach and the implementation code, as illustrated in previous study [14], consists of four basic sections. Phase change and material removal will be illustrated from the MD retrieved data. The Morse Potential Function is being used for the description of the inter-atomic potential and the potential energy within the system’s particles. The initial positions of the atoms are determined according to the BCC lattice and the initial velocities are randomly ascribed to the atoms according to the Maxwell–Boltzmann distribution at room temperature.

Assuming that the target is infinitely large, in the lateral directions, the Periodic Boundary Conditions (PBC) were applied. The top surface of the target, where the laser beam irradiates, has been subjected to the Free Boundary Conditions (FBC) allowing particles to move and be removed. At the bottom surface, the Reflecting Boundary Conditions are applied (RBC) to ensure velocity damping. The laser beam has a Gaussian profile in time and space [15]. However, it is the combination of the uniform distribution in space and the Gaussian distribution in time that is preferred [16]. A number of photons corresponding to the energy produced by the laser are deposited into the bulk material target. The deposition is exponential and follows the Beer Lambert law. By absorbing photons, the particles of the crystal increase their kinetic energy and consequently, their velocity is also increased. Hence, their new velocities have to be recalculated following their irradiation. The particle removal criterion is based on the cohesive energy, one of the most significant factors in an MD simulation, as it determines the amount of energy that can be absorbed by an entity.

2.2. Simulation Details

Iron (Fe) is being selected as the metal with a very broad industrial application. The parameters of Fe used in the simulation model are shown in Table 1 and are assumed to be constant during the simulation time intervals.

| λ (nm) | R (%) | Tm (K) | Tv (K) |
|--------|-------|--------|--------|
| 775    | 65    | 1811   | 3134   |

The MD simulation system region is formed from BCC unit cells, in the dimensions of 3.8x3.8x726.5nm [10446.3nm³] and consists of a total number of 719,756 particles. The velocity Verlet algorithm has been applied to the system for integrating the equations of motion. The interaction among the atoms in the system is described in the Morse Potential Function. According to this potential, the Morse parameters of Fe are computed from the energy of sublimation, the lattice constants and the compressibility for a perfect lattice are shown in Table 2.

| D (eV) | a (Å⁻¹) | e₀ (Å³) |
|--------|---------|---------|
| 0.4174 | 1.3885  | 2.845   |

The initial velocities are randomly ascribed to the particles according to the Maxwellian distribution at

Fig. 1. Schematic of the problem description
300K room temperature. The ablation is investigated up to several tens of femtoseconds. The Periodic Boundary Conditions (PBC) have been applied, in the X and Y directions of the computational domain, for simulating an infinite medium. A velocity dampening technique is applied, at the bottom of the computational cell along with the RBC, in order to prevent the artificial ablation effects that may arise from the shock wave reflection. The number of photons, corresponding to the laser energy, is deposited in the material exponentially following the Beer-Lambert law. The laser wavelength is set at 800 nm. The pulse duration, the total interaction time of the laser and the material are fixed for all experiments, whilst the laser fluence ranges from 0.1 J/cm² to 30 J/cm².

3. Experimental Setup

For the verification of the model, a set of femtosecond laser ablation experiments was conducted. The laser system used in the experiments consisted of a mode-locked Ti:Sapphire laser system (CPA-2101, Clark-MRX, Inc) pumped with a CW, solid state, Nd:YVO4 laser (Millenia Vs, Spectra Physics). This system is tunable from 400 to 800 nm and provides ultrashort laser pulses in the femtosecond regime (duration < 100 fs). A 5-fold expanding telescope was used for expanding the laser beam in order to uniformly illuminate the back-aperture of an oil immersion objective lens (100x; N.A., 1.2). This tight focusing geometry has made available the high fluences, required for the ablation of the samples. The irradiation power was controlled with a combination of a motorized λ/2 plate and an ultrafast polarizer (Melles Griot, 16PPB200) and was measured at the back aperture of the lens used for excitation. To control the desired total interaction time of the laser beam with the material target, a fast mechanical shutter (rise system time 3ms, Newport 845HP) has been integrated into the setup.

![Table 3. Parameters of the laser system](image)

| Laser Type | Wavelength (nm) | Pulse Duration Range (Fs) | Spot Diameter (μm) |
|------------|----------------|--------------------------|--------------------|
| Ti: Sapphire Laser | 400-8000 | 50-80 | 3134 |

The Scanning Electron Microscope (SEM) is used for observing the ablated area while the laser Scanning Microscope (LSM) for measuring the ablation depth. The ablation depth per pulse is evaluated by measuring the total depth of the drilled hole and divided by the number of pulses used.

![Table 4. Properties of Fe Samples](image)

| Material | R % | α (cm⁻¹) | 1/α (nm) | Tm (K) | Tc (K) |
|----------|-----|----------|----------|--------|--------|
| Fe       | 99.99% | 65 | 0.54 x 10 | 18.5 | 1811 | 3145 |

4. Discussion of the Results

The ultrashort laser ablation has been investigated computationally and experimentally in the range of laser fluences from the threshold up to 10 J/cm². Both computational and experimental results define the different ablation regimes of an ablation threshold.

4.1. Ablation Depth and Laser Fluence

The ablation depth, per laser pulse, has been used as a validation means of the theoretical model. Figure 2, presents a comparison of the ablation depth in the experimental and computational results. For the majority of the laser fluences applied, there is observed a good agreement of the results.

![Fig. 2. Computation and experimental ablation depths vs applied laser fluence](image)

Starting from the lowest applied laser fluence up to the value of 0.1 J/cm², the ablation depth per pulse, follows a linear increment with the corresponding fluence, as shown in Figure 3. For the experimental results the smallest measured depth is 0.5nm for a 0.07 J/cm² pulse. This is due to the fact that the LSM does not provide accurate measurements for depths smaller than 25 nm. However, the SEM images of the specimens
show material removal from the surface for fluences even smaller than 0.07 J/cm². Computational results indicate that the ablation phenomenon starts at 0.06 J/cm².

Fig. 3. Computation and experimental ablation depths vs applied laser fluence for the first ablation region from 0.01 to 0.1 J/cm²

The second distinct ablation area in relation to the applied laser fluence varies in-between 0.1 and 1 J/cm² as shown in Figure 4. Ablation starts to become intense, producing depths that start from 2 nm to 12 nm per pulse. The ablation depths show an exponential growth in relation to the laser fluence applied to both the computational and experimental results.

Fig. 4. Computation and experimental ablation depths vs applied laser fluence for the second ablation region from 0.1 to 1 J/cm²

In-between the limits of 1 and 10 J/cm² lies the third ablation area. The ablation process is intense, resulting in the production of holes whose depths vary from 12 nm to 500 nm per pulse. The ablation depths continue having an exponential growth in relation to the laser fluence applied to both computational and experimental results.

Fig. 5. Computation and experimental ablation depths vs applied laser fluence for the third ablation region from 1 to 10 J/cm²

The accuracy of the MD model is verified by comparing the ablation depth with the experimental data obtained. Both experimental and theoretical results clearly define three (3) different ablation areas, each governed by different ablation mechanisms. At laser fluences, near the ablation threshold (0.1 J/cm²), the removal of the material is governed by the photothermal mechanism. Increase in the fluence results in photomechanical mechanisms responsible for the material’s removal and in thermo-elastic stresses being developed due to fast heating. Here, the ablated material starts being composed of big clusters. The increase of the fluence results in a strong overheating of the absorbed volume and in a subsequent phase explosion, while it leads to the decomposition of the ablated material into liquid droplets and single particles. At the ablation areas, there is a transitional limits co-existence of more than one ablation mechanisms being responsible for the material’s removal [18]. Figures 3 to 5, show the dependence of the ablation depth per pulse on the Laser fluence. There is a good agreement between the MD and the experimental values. The discrepancy observed could refer to the lack of knowledge of the reliable data for the thermo-physical parameters of the electron system.

4.2. Laser Ablation Efficiency

As it is defined in chapter 1, laser ablation efficiency is considered the ratio of the ablated material volume to the energy of the laser pulse. The diagram of Figure 6 can be divided in three areas based on the behavior of the laser efficiency. The first area is up to laser fluence 1 J/cm². Within this area the laser efficiency presents a linear increase of a small slope. The second area has a range from 1 J/cm² up to 10 J/cm². In contrast to the behaviour of the laser
efficiency in the first area, within the second area the growth of the laser efficiency is exponential. The last area is dedicated for laser fluence after the value of 10 J/cm². Within this area the laser efficiency follows an exponential decrease. It is evident that the laser fluence region around 8-10 J/cm² present the highest values of laser efficiency. Thus the laser fluence of 10 J/cm² can be considered as a threshold for laser efficiency that it should not be transgressed, since after this value the laser efficiency only decreases.

The explanation of the laser efficiency decrease can be found in four main reasons. Firstly, shock waves appear and affect significantly the mechanism of the laser ablation after the laser fluence increase. Secondly, the material volume shrinks due to the high pressures occurred by the shock waves. Ejected atoms or molecules ionized facilitating the plasma or plume creation consisting of free electrons and ions. Finally, plasma detachment reduces the absorption properties of the material leading to a further laser efficiency decrease.

Fig. 6. Laser ablation efficiency as a function of the laser fluence

5. Conclusion

This study presents results of the MD modelling and experimentation with femtosecond laser pulses, regarding the ablation mechanisms Fe, at fluences ranging from 0.01 up to 10 J/cm² and emphasizes on the laser efficiency. The theoretical results produced by the MD analysis are in agreement with the experimental results. Three main ablation areas are identified, the first ranges from 0.01 up to 0.1 J/cm², the second from 0.1 to 1 J/cm² and the last one from 1 to 10 J/cm². The ablation phenomenon based on computational results that are in line with theoretical is initiated around 0.06 J/cm². The ablation efficiency behaviour is divided into three areas, and the maximum ablation efficiency is identified in the area of 8-10 J/cm².

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