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The influence of double pulse delay and ambient pressure on femtosecond laser ablation of silicon

Ying Qi, Hongxia Qi, Qinxin Wang, Zhou Chen, Zhan Hu *

Institute of Atomic and Molecular Physics, Jilin University, Changchun 130012, People's Republic of China

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The effects of double pulse delay time and ambient pressure on femtosecond laser ablation of Si are studied by means of photoluminescence and morphology analysis. Detailed scan of double pulse delay is performed at vacuum (2 Pa) and atmospheric pressure, and detailed scan of pressure is performed at three double pulse delay times of 0.2 ps, 53.13 ps and 106.47 ps. It is found that, at various fluences, photoluminescence intensity and morphology change as functions of both double pulse delay and ambient pressure. Especially, the shape of the splashing droplets also changes at different experimental conditions, indicating higher or lower sample temperature. The observations are explained by the efficiently energy coupling between the second pulse and the liquid layer produced by the first one, and the pressure dependent energy coupling between plasma and liquid phase as well as ambient gas.

1. Introduction

Laser ablation of material is a complex process involving several phases and multiple length and time scales [1–3]. A sequence of different events is initiated after the laser irradiation. The type and order of these events are determined by the properties of the material and the laser [4,5]. Micromachining with laser pulses of femtosecond width has been shown to limit the amount of thermal diffusion due to the very short heating duration and the rapid solid-to-vapor phase transition. The absorption region is limited to penetration depth of the optical pulse; thus collateral damage is limited.

Double pulses have been used to probe or control the ablation mechanisms, such as energy deposition, heat transfer, material removal processes in various materials of metal, semiconductor or dielectrics [6–8]. It is shown that photoluminescence emission during ablation is enhanced when the double pulse delay, defined as the time between two sub-pulses, is set to a few tens of picoseconds [6,7]. Compared with nanosecond and picosecond laser pulses, the plasma reheating effect can be ignored in the femtosecond single pulse ablation process [9–13]. However, with femtosecond double pulses, plasma reheating does show some effect. For example, at the fluence of 15.1 J/cm^2, in copper ablation experiments [14], it is observed that the plasma luminosity increases with double pulse delay, while the ablation depth decreases. These results reveal that the interaction of laser–plasma plays a prominent role during the whole ablation process, and the results are in qualitative agreement with the simulation result described in Ref. [15]. With the laser fluences of 1.6 kJ/cm^2 and 1.2 kJ/cm^2, maximum enhancement of ion density and average ion energy in ablation plume of silicon is observed at the double pulse delay time of 5 ps [16,17]. The results are explained by the appropriate plasma density gradient with a properly positioned critical surface, which interacts most strongly with the second pulse.

The effect of the surrounding gas and pressure on laser ablation of solid sample has received much increased attention in recent years because of its importance in pulsed laser deposition [18], nanoparticle formation and growth [19], laser micromachining [20], and laser-induced breakdown spectroscopy (LIBS) [21]. After the irradiation of the surface of solid sample by high powered laser pulse, the produced plasma expands as a shock wave into the surrounding atmosphere at supersonic speed. The energy of plasma transfers into the environment gas through several processes [22,23], such as shock wave heating, thermal conduction, radiative emission, and ion recombination. The selection among these interaction processes depends on the pulse energy, pulse duration, laser wavelength, as well as composition and condition of environment gas. With nanosecond laser pulse, Lobe et al. studied the influence of the environment gas and gas mixture on the ablation of steel [24]. The ablation efficiency can be increased not only by increasing the laser fluence, but also by changing the ambient pressure. The results are attributed to the enhanced interaction between the plasma and the sample in certain ambient gas pressure. Bashir et al. [25,26] analyzed the influence of the different gases at the different pressures on optical emission intensity, electron temperature and density. It is revealed that nanosecond LIBS performance is strongly affected by the ambient condition due to the shielding effect of plasma. For femtosecond laser ablation [27–29], at a certain background air pressure (around 500 Pa), the photoluminescence intensity

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* Corresponding author.
E-mail address: huzhan@jlu.edu.cn (Z. Hu).

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can be significantly enhanced compared with the atmospheric pressure, which is attributed to the further expansion of plasma into ambient gas and not being cooled as rapidly by the surrounding species. While at low pressure condition (around 0.4 Pa), the decreased emission intensity is due to the decreased collisional excitation and less energy transmission between the plasma and the surrounding gas particle. In addition, more thermal energy is retained in sample under gas environment, compared to lower pressure condition as reported in [30–32], because the high-pressure plasma can affect the coupling of the thermal energy with the sample. The explanations of the results are based on two mechanisms: a certain amount of the laser energy stored in the plasma can be transferred to the sample surface; compared with vacuum condition, the increased redeposition of the ablated material at higher gas pressure enhanced the thermal energy coupling to the sample. In order to explain the enhancement of retained thermal energy in a gas environment, a combined model is developed to simulate the ablation process, which is based on 2D thermal model of laser-induced heating and ambient gas dynamics [33]. The simulation shows that the hot plasma moves preferentially toward the sample and its thermal energy couples efficiently with the sample. Time-resolved pump-probe shadow graphic imaging results also show that the ambient air assisted in dissipating the energy into the sample [34].

Silicon and its compounds are widely used in semiconductor industries, the properties of which and their interaction mechanisms with external fields have been the study subject of many experimental as well as theoretical works [35–37]. The interaction of Si with laser has received more and more interest [38,39]. In our previous studies [6,7], we also used Si as the sample, which is ablated by femtosecond double pulse at very low fluences and atmospheric pressure. It was found that, with increasing double pulse delay, photoluminescence intensity increases monotonically. However, the enhancement ratio of photoluminescence intensity at longer double pulse delay to that at shorter delay will decrease, with increasing fluence. The ablation behavior under higher laser fluence will be studied in this paper. It was also observed in our previous work that, with increasing double pulse delay, the amount of resolidified liquid material increases. However, the crater volume does not change much with double pulse delay, which was attributed to the redeposition of materials back into the crater. Under lower environment pressure condition, the behavior of ejected material may be quite different. So the study of ablation under different environment pressure conditions is important.

In this paper, in order to further reveal the mechanisms of femtosecond laser interaction with materials, we investigate systematically the ablation of Si with double femtosecond laser pulses. At different double pulse delays and ambient pressure values, besides photoluminescence intensity, the morphology of ablation craters (including splashing droplet shape, crater depth, width and volume) is studied, which was not reported in other works. Especially, we do careful analysis of the splashing droplet shape and show that the shape changes under different experimental conditions, which reflects the ablation mechanisms. A complete scan of all double pulse delays and pressure values is not possible. We do detailed double pulse delay scans at vacuum condition (2 Pa) and atmospheric pressure, which will be discussed in Section 3.1; and then do detailed pressure scans at three double pulse delay values of 0.2 ps, 53.13 ps, and 106.47 ps, which will be discussed in Section 3.2.

2. Experimental

The experimental setup used in this paper is similar to that described in Ref. [6], as shown in the schematic drawing in Fig. 1, which consists of a femtosecond laser source, a Michelson interferometer to generate double pulse with a controllable delay, a sample manipulator, and an optical detector. A regenerative amplifier laser system (Spectra Physics Tsunami oscillator and Spitfire amplifier) producing 800 nm, 110 fs (FWHM) pulses at a repetition rate of 1 kHz with pulse energy up to ~0.7 mJ is used. Laser energy is varied by adjusting the half-wave plate followed by a Glan prism. The laser is then divided into two beams and later combined into one beam in a Michelson interferometer setup, in which the optical length of one beam can be adjusted, resulting in a varying double pulse delay from −53.53 ps to 106.47 ps. For all the experiments the intensities of the two pulses are equal. The overlapped pulses are focused onto the sample by a 10× objective lens (Edmund Optic, NT59-877) with a numerical aperture of 0.28, and working distance of 33.5 mm. The focus size is measured using a moving knife-edge method [40] to be 0.97 μm in diameter, which results in 135 J/cm^2 fluence with a 1 μJ pulse energy. The sample is undoped Si$\langle 111 \rangle$ wafer (MTI Crystal), which is mounted on a motorized XYZ high precision translation stage (Newport, M-462-XYZ-SD stage and TRA25cc actuators) in a vacuum chamber. The sample's normal axis is at an angle of 30° with respect to the laser beam direction. The laser is operated in single shot mode and the stage moves the sample to a new location after each laser spot. Photoluminescence perpendicular to the laser beam is collected by an *f*/2.0 lens, focused into a spectrograph (Princeton Instruments, PI Acton SP500i) and detected by an ICCD detector (PI-MAX, 1024 × 256 pixel). The ICCD is operated in shutter mode with an exposure time of 100 ms which is long enough to collect any photoluminescence generated. The spectrum intensity at all wavelengths in the spectrum range studied (350–600 nm) follows the same trend as a function of double pulse delay. So in this work, the strongest Si$^+$ emission line at 505.6 nm, from $3s^2 4p(^2P_{3/2}) \leftarrow 3s^2 4d(^2D_{5/2})$ transition, is measured. An average of typically 25 shots is taken for each data point. When we investigate the effect of ambient pressure during the ablation process, air is used as buffer gas. The morphology of the ablated crater is measured by an Atomic Force Microscope (AFM, BenYuan, CSPM5500) operated in contact mode. The maximum lateral resolution and vertical resolution of AFM are 0.2 nm and 0.01 nm, respectively. The scanning range is 50 × 50 μm.

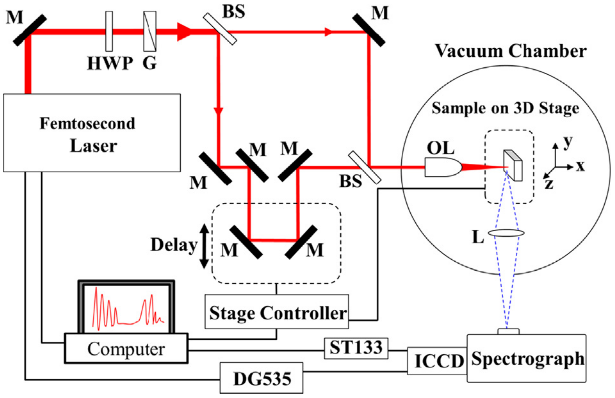

Fig. 1. Schematic drawing of the experimental setup. The setup can be divided into four parts: laser, laser beam manipulation, vacuum chamber, control and data acquisition. Laser beam manipulation components include half-wave plate (HWP), Glan prism (G), beam splitters (BS), high reflective mirrors (M), and microscope objective lens (OL). Control and data acquisition components include 1D (Delay) and 3D stages, lens (L), stage controller, DG535 delay generator, ST133 camera controller, intensified CCD camera (ICCD), spectrograph, and computer.

3. Results and discussion

3.1. Double pulse ablation of Si with different delay values and laser fluences in vacuum and atmospheric pressure

First, we ablate the Si sample in vacuum (2 Pa) and atmospheric condition with different laser fluences and double pulse delays. The photoluminescence obtained in atmospheric condition and in