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2019

Farrokhi, H., Gruzdev, V., Zheng, H., & Zhou, W. (2019). Fundamental mechanisms of nanosecond-laser-ablation enhancement by an axial magnetic field. *Journal of the Optical Society of America B*, 36(4), 1091–1100. doi:10.1364/josab.36.001091

<https://hdl.handle.net/10356/144719>

<https://doi.org/10.1364/JOSAB.36.001091>

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Fundamental mechanisms of nanosecond-laser-ablation enhancement by axial magnetic field

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Steady magnetic field perpendicular to laser beam is widely used to improve rate and quality of laser ablation. Recently we reported a 69-fold enhancement of laser ablation of silicon by magnetic field parallel to laser beam. To understand the fundamental mechanisms of that phenomenon, multi-pulse magnetic-field-enhanced ablation of stainless steel, titanium alloy, and silicon was performed. The influence of the magnetic field significantly varies depending on the material: from 2.8-fold ablation enhancement on stainless-steel and silicon to no pronounced ablation modification on titanium alloy. Those results are discussed in terms of magnetized-plasma, magneto-absorption, skin-layer, and magnetic-field-influenced transport effects. Understanding of those mechanisms is crucial for advanced controlling of nanosecond laser-surface coupling to improve laser micromachining.

OCIS codes: (140.3390) Laser materials processing; (350.3390) Laser materials processing; (160.6000) Semiconductor materials; (160.3900) Metals.

1. INTRODUCTION

Laser ablation is a laser interaction with a solid surface by which laser radiation removes matter from the surface [1-3]. In that entire research field, ablation by nanosecond laser pulses has received the most significant attention because of a broad range of industrial applications to precisely micro-machine various materials [2, 3]. One of the major challenges for advancing the nanosecond-pulse laser ablation and micro-machining is to optimally balance laser-surface coupling. Coupling of higher laser-pulse energy increases the rate of material removal, but the quality of the laser-machined structures degrades because of significant collateral damage and contamination of the surface by intensive thermal effects [4, 5]. Reduction of laser-pulse energy results in the improvement of machining quality but leads to a lower speed and throughput of material processing. Therefore, a fundamental approach is required to enhance material removal without increase of laser-pulse energy to improve the balance between speed and quality of the nanosecond laser ablation. Stimulated by this demand, extended basic and applied research has been carried out to address the

fundamental issues of a reliable control over the laser-surface coupling [2-24].

Among those issues, shielding effect by laser-produced plasma (LPP) is one of the most significant. The plasma reduces laser-surface coupling by absorbing and reflecting a significant part of ablating laser pulse if its duration is longer than the time of plasma formation. Because of this fundamental reason, reduction of the LPP shielding effect is among the major approaches for improvement of the laser-surface coupling for nanosecond pulses [5-12]. Steady magnetic field [8-19, 21], ablation under vacuum conditions [7, 20], and *dc* electric field [21, 22] are employed to remove the LPP from a laser beam, to confine LPP to a smaller volume, or to lower plasma density. Of those approaches, use of steady magnetic field has received the most attention due to the feasibility of a better, more flexible, and more stable control over the laser-surface coupling [8-19].

The fundamental mechanism of the magnetic-field-supported reduction of plasma shielding is associated with the action of the magnetic Lorentz force $\mathbf{F}$ [8-19]:

$$\mathbf{F} = q (\mathbf{v} \times \mathbf{B}), \quad (1)$$

where q is electric charge of a particle moving at speed $\mathbf{v}$. To produce the largest force, magnetic field $\mathbf{B}$ should be aligned parallel to ablated surface and normal to the laser beam to maximize the vector product of $\mathbf{B}$ and the speed $\mathbf{v}$ of ablated matter (Fig. 1(a)). This alignment of the magnetic field increases the ablation rate by a factor of 1.5– 2.5 and reduces debris deposition when applied to surfaces of paramagnetic conducting materials [8-12]. It is also employed in studies of the LPP properties and confinement by the magnetic field [13, 14, 17-19]. Alignment of the magnetic field parallel to the laser beam (also referred to as axial) has received minor attention [15, 16] because the ablated-matter speed $\mathbf{v}$ is almost parallel to the magnetic field and minimizes the Lorentz force (Fig. 1(b)). Due to this reason, this alignment is considered as not favorable for an effective LPP confinement [8-19].

Fig. 1. The traditional longitudinal alignment of the magnetic field (blue arrows), i.e., parallel to a surface and normal to laser beam (a) delivers a larger Lorentz force $\mathbf{F}$ due to almost normal alignment of the magnetic field ($\mathbf{B}$) and speed ($\mathbf{v}$) of the particles 1 and 2 departing from the surface. The axial alignment of the magnetic field (b) produces a significantly smaller Lorentz force.

However, improvement of the laser-surface coupling can be approached via a magnetic-field-induced modification of material response to the laser action rather than by confining the LPP. For example, experiments on paramagnetic metals like tin have shown some 2-fold increase of ablation rate by the axial magnetic field [17]. Recently reported 69-fold enhancement of ablation of silicon by nanosecond UV laser pulses [23, 24] confirms the feasibility of even stronger intensification of ablation by the axial magnetic field. Those results are not justified by the LPP confinement [15] because of minimizing of the vector product ($\mathbf{v} \times \mathbf{B}$) by the axial alignment. A brief

analysis of possible mechanisms of the ablation enhancement [23, 24] suggests contributions from propagation through the magnetized plasma, modified dynamics of debris deposition, magneto-hydrodynamics of melt motion, and modification of optical response of silicon. However, the axial magnetic field modifies a broader range of material properties involved in the ablation processes, e. g., heat conductivity. Therefore, more consideration should be given to the phenomena associated with the magnetic-field-induced modification of those material properties.

Motivated by those reasons, here we report experiments on ultraviolet (UV) nanosecond multi-pulse ablation of silicon, stainless steel, and titanium alloy under zero and 0.341 T axial magnetic field. Highly different levels of ablation enhancement are obtained under the same magnetic field depending on the type of ablated material. Qualitative consideration of absorption and transfer of laser-pulse energy suggests that modification of transport properties of the solids by the axial magnetic field significantly contributes to the observed ablation enhancement. The reported results also confirm that the ablation enhancement is material specific and is not equally effective on arbitrary solids. The rest of this paper is organized as follows. Section 2 contains a description of the experimental setup and ablation procedure. Section 3 summarizes the major results of the experiments. Section 4 contains the extended discussion of the experimental results, analysis of the physical mechanisms, and estimations to support the conclusions drawn in Section 5.

2. EXPERIMENTAL SETUP AND DETAILS

The samples used in the experiment were a single crystal n-type silicon wafer (phosphorous doped; surface $<100>$; thickness 300 μm), and polished flat samples of stainless steel (316L) and titanium alloy (Ti6Al4V). Each sample was cut into 10×10 mm² pieces. Ultrasonic cleaning of the samples was carried out in methanol for 10 min, and then in de-ionized water for another 30 min. 20-ns pulses with a Gaussian beam profile were generated by a Q-switched third-harmonic Nd:YVO₄ laser at wavelength 355 nm and repetition rate 1 kHz. Experiments were conducted in air. The mean laser spot diameter was 51 μm (at FWHM of intensity). To generate a uniform axial magnetic field of 0.05 - 0.4 T, a custom-made copper coil was powered by *dc* current (see Fig. 2). A sample was mounted deep inside the coil to provide a uniform magnetic field normal to the sample surface and parallel to the laser beam. To make a groove, the entire assembly composed of the copper coil, the stage, and the sample was placed on a motorized XYZ stage and moved under the laser irradiation (see Fig. 2). Ablated samples were imaged with scanning electron (SEM) and 3D optical microscope to characterize depth, width, and general morphology of the ablated structures.

Several grooves on each of the samples were ablated by moving the stage at the speed of 1 mm/sec. The regime of the ablation was essentially multi-pulse since a few tens of pulses interacted with each site of the laser-beam trajectory on the surface. The experimental procedure was

as follows: on each sample, a groove was produced without the magnetic field; then the laser beam was shifted by a few millimeters in the direction normal to that groove, the coil was powered to produce required magnetic field, and another groove was made parallel to the first one under assistance of the magnetic field. Value of the magnetic field was kept the same in all experiments on the metals to deliver consistent results for interpretation.

Fig. 2. Sketch of the experimental setup for axially magnetized nanosecond laser ablation and a scheme of the mutual alignment of the laser beam and laser produced plasma (LPP) under the axial magnetic field.

3. RESULTS

A few grooves were produced on the silicon surface by 35- μJ laser pulses (peak fluence is 1.19 J/cm²). Typical results are shown in Fig. 3. The laser fluence was close to the ablation threshold of silicon at zero magnetic field [23] and produced nano-grooves with average depth 142 nm (Fig. 3(a) and (c)). However, when the magnetic field of 0.341 T was applied directed towards the surface, the groove depth increased to 9.8 μm , i.e. by a factor of 69 at the same amount of pulse energy and fluence (Fig. 3(b) and (d)). Also, the magnetic field increases the melt rim at the edges of the groove. Among other remarkable results is the periodic cyclic-type perturbation of the laser track by the magnetic field with a period about 35 μm (Fig. 3(b)).

Several single spots were ablated on the silicon surface at higher fluence (pulse energy is 112 μJ ; peak fluence is 3.81 J/cm²) by applying 10 [23] and 30 laser pulses per spot. Remarkable reduction of debris deposition by the magnetic field was observed [23]. Depth of the ablated craters increased from 10 (with no magnetic field) to 37 μm (with 0.192 T magnetic field) for ablation by 10 pulses, and from 37 (with no magnetic field) to 90 μm (with 0.192 T field) for ablation by 30 pulses. Further details of the experiments on the silicon sample and characterization of the laser-ablated structures can be found in Ref. 26.

The grooves produced on the stainless-steel surface by 112- μJ laser-pulses with no magnetic field showed large amounts of the melt at the rims of the groove (Fig. 4(a)). An average depth of the grooves was about 13 μm (Fig. 4(c)). Axial magnetic field of 0.341 T directed towards the surface increased the groove depth to 35 μm . Remarkable modification of morphology of the melt deposits without appreciable increase of the melt rim was observed at the edges of the groove produced under the magnetic field (Figs. 4(b) and 4(d)). Characterization of the groove profiles uncovered that the magnetic field substantially improved the aspect ratio of the grooves from 0.34

(13/38) to 1.52 (35/23). A similar amount of increase of the aspect ratio by reduction of diameter/width of the laser-ablated structures was observed on silicon when ablated by 112- μJ pulses [23]. In contrary to silicon, the magnetic field did not produce a periodic cycling perturbation of the grooves produced on stainless steel.

Fig. 3. SEM images (a, b) and depth profile (c, d) of ablation area produced on silicon without (a, c) and with the magnetic field (b, d) directed normally towards the surface.

Fig. 4. SEM images (a, b) and depth profile (c, d) of ablation area produced on stainless-steel without (a, c) and with the magnetic field (b, d) directed towards the surface.

Finally, a linear scanning of the laser beam was performed on the titanium-alloy surface at the pulse energy of 112 μJ (Fig. 5). The laser action produced irregular roughness with the domination of linear structures (Fig. 5(a) and 5(c)). Significant debris was observed at the edges of the ablated area. When the axial magnetic field of 0.341 T was applied, the roughness structure dramatically changed (Fig. 5(b)), but no significant increase of ablation depth was observed (Fig. 5(d)). A remarkable feature of the magnetic-field-assisted ablation is a very pronounced reduction of contamination and debris deposition at the edges of the ablated area (Fig. 5(b)).

Fig. 5. SEM images (a, b) and depth profile (c, d) of ablation area on titanium-alloy surface treated without (a, c) and with the magnetic field (b, d) directed towards the surface.

4. DISCUSSION

Silicon and stainless steel showed qualitatively similar behavior: there was an evident ablation without magnetic field; the axial magnetic field increased ablation depth, improved aspect ratio, and reduced debris deposition. Titanium alloy behaved in a qualitatively different way and showed significant increase of surface roughness within a laser track without formation of any appreciable ablated structure like a groove. Considering the potential contributions of various physical effects, we focus on those that can explain this qualitative difference.

First, we note that parameters of the nanosecond UV laser pulses are favorable for thermal ablation that runs under a quasi-equilibrium energy transfer [2-4]. The thermal ablation is attributed to a variety of coupled phenomena [1-4]. For interpretation of the experimental data, we consider four groups of the phenomena: (a) laser-plasma interactions including propagation through magnetized LPP; (b) laser-surface coupling including the effects attributed to reflection, penetration, and

absorption; (c) transport effects that include transfer of absorbed laser energy through both liquid and solid phases; (d) other effects that include thermo-mechanical stresses; material removal from the surface, e. g., evaporation; melt hydrodynamics; and surface oxidation. The magnetic field can influence all these effects. To evaluate the influence for each of the ablated solids, we do estimations based on the material parameters summarized in Table 1 to identify the processes and material properties that can make a major contribution to the reported above ablation enhancement by the axial magnetic field.

A. LASER-PLASMA INTERACTION EFFECTS

Plasma magnetization can contribute to the enhancement of the laser-surface coupling in several ways. The key parameters to evaluate the magnetization effects are the fastest rate of LPP confinement; modification of LPP optical response; and modification of LPP extension by the magnetic field.

The rate of the LPP confinement by the axial magnetic field is characterized by cyclotron frequency or period of the plasma particles [8-19, 23]:

$$\omega_c = \frac{qB}{m}; \quad \tau_c = \frac{2\pi}{\omega_c} \quad (2)$$

where m is mass of a charged particle of the LPP; q is a particle charge. Among the heavy-mass components of the LPP, the highest cyclotron frequency should be attributed to a single-charged ion according to Eq. (2). The cyclotron frequency is of the order of 10^6 Hz for a single-charge silicon ion. Atomic masses of most components of the metals are almost two times larger than that of silicon (Table 1). Correspondingly, their cyclotron frequencies are two times lower. Therefore, the heavy components of the LPP produced by ablation of the metals are confined approximately two times slower than in case of silicon. The low-mass LPP component consists of electrons, which cyclotron frequency is 6×10^{10} Hz for all the materials.

Table 1. Material parameters and optical properties at wavelength 355 nm.

Material	Reflectivity [%]	Absorption coefficient [$\times 10^5$ 1/cm]	Penetration depth (estimated from absorption) [nm]	Electron mean free path [nm]	Atomic mass [atomic units]	Melting/Boiling points [K]	DC resistivity [Ohm m] (at 298 K)
Si (crystal)	56.16 [27]; 57.25 [28]	10.13 [27]; 10.34 [28]	9.87; 9.67	20 -60 [29]	28.09 (Si) 30.93 (P)	1687/2628 [33]	2.3×10^3 [34]
Stainless steel (316L)	45.4 [25]; 68.45 [26]; (iron)	8.34 [25]; 11.54 [26];	11.99; 8.67;	7.8 - 9.0 (estimated from resistivity)	55.85 (Fe) 52.00 (Cr) 58.69 (Ni) 54.93 (Mn)	1700 (steel) / 2870 (iron) [36, 37]	0.75×10^{-6} [35]
Titanium alloy Ti6Al4V	55.0 [25]; 52.5 [26] (Ti)	10.17 [25]; 8.61 [26]	9.84; 11.61	2.9 - 3.9 [30-32]	47.88 (Ti) 26.98 (Al) 50.94 (V)	1922 / 3290 (Ti) [36, 38]	1.7×10^{-6} [38]

Variation of the LPP linear optical response by the magnetic field is evaluated by the Appleton formula for the dielectric response of a magnetized plasma [23]. The reported above magnetic field produces a negligible modification of the plasma optical response since the ratio of the cyclotron frequency to laser frequency is of the order of 10^{-10} for the heavy LPP components and about 10^{-5} for the LPP electrons [23]. Therefore, the linear laser propagation through the magnetized plasma is not appreciably affected by the magnetic field.

The magnetic-field-induced changes of LPP expansion can be characterized in terms of Larmor radius of the LPP particles. Following our previous analysis [23], we assume the initial speed of the LPP heavy particles to be the average thermal speed at the boiling point T_{BOIL} :

$$R_L = \frac{m\bar{v}}{qB} = \frac{\sqrt{2k_B m T_{BOIL}}}{qB}; \quad \bar{v} = \sqrt{\frac{2k_B T_{BOIL}}{m}} \quad (3)$$

where k_B is the Boltzmann constant; $\bar{v}$ is average particle speed. Estimations by Eq. (3) show that the Larmor radius of single ions of silicon LPP is about 45-50% smaller than that of stainless steel and titanium alloy. This result suggests debris back-deposition over a larger area in case of the metals compared to silicon. Although it is favorable for debris reduction, it can hardly explain the magnetic-field-induced increase of the ablation depth.

Those simple estimations suggest a negligible influence of the LPP magnetization on the ablation rate, but other effects can make appreciable contributions. First, emission of LPP can enhance heating of the ablated surface since the LPP emission adds 10-20% to the direct laser-induced heating of the surface [39]. Moreover, that contribution substantially depends on the type of ablated material [13, 14, 17-19] and can partly justify the different ablation behavior of the three materials. Although the influence of the axial magnetic field on the LPP emission has not been studied in detail, enhancement of the LPP emission by a longitudinal magnetic field [13, 14, 17-19] suggests a similar effect may take place in the axial magnetic field. Second, self-focusing of the laser beam can happen in the magnetized plasma due to its nonlinear optical response. Since the nonlinear response enhances with the increase of plasma-density gradient [39], localized confinement of the LPP within a laser spot and its high density (especially at the early stages of expansion) may result in a remarkable amount of the LPP optical nonlinearity and appreciable Kerr-type lensing effect. This effect is material-specific and may contribute to the observed material dependence of the ablation via two mechanisms: (a) dependence of the nonlinear optical response on nature and density of atoms and ions constituting the plasma; and (b) dependence of ablation rate and LPP density on properties of ablated materials. Third, the LPP is a complex or dusty plasma [41] since it contains neutral atoms as well as charged and neutral clusters of atoms [13, 14, 17-19]. The complex plasmas subjected to the action of moderate magnetic fields can exhibit specific dynamics due to the magnetization of the atomic clusters [41]. This effect can also be influenced by the composition and properties of the ablated materials. Therefore, the magnetization of LPP by the axial magnetic field can affect

the ablation rate due to variation of the laser-surface coupling by those non-trivial physical effects. Evaluation of those LPP contributions requires extended simulations of the laser-plasma interactions that go far beyond the scope of this article.

B. LASER-SURFACE COUPLING

All the three materials are good absorbers at laser wavelength of 355 nm [25-28] and reflect slightly more than 50% of incident light (Table 1). This fact assumes the amount of laser-pulse energy coupled into the materials without the magnetic field is approximately the same. The thickness of a subsurface layer where the absorption happens is characterized by penetration depth δ_n . Evaluated as inverse absorption coefficient, the depth is about 10 – 11 nm for all the materials at zero magnetic field (Table 1). Therefore, the zero-magnetic-field conditions of the laser-surface coupling are very similar for all the three materials, and the strikingly different response of the titanium alloy is driven by other effects.

The penetration depth is affected by several processes [42] that are differently influenced by the magnetic field. The major of them are absorption by free carriers (that are electrons in all the materials under consideration), absorption due to inter-band electron transitions, and absorption due to collective free-carrier excitations, e.g. plasmon-polaritons. The collective excitations cannot be effectively generated by the laser pulse in the metals because their plasma frequency [42] is larger than the laser frequency. In silicon, the conduction-carrier density is not sufficient to support any appreciable plasmon-polariton effects at laser wavelength at the beginning of a laser pulse. However, generation of free electrons by the laser-driven inter-band excitation increases the free-electron plasma frequency and makes it equal to the laser frequency at conduction-electron density about $8.1 \times 10^{21} \text{ cm}^{-3}$ (evaluated for longitudinal effective mass at X-valley of the silicon conduction band [42, 46]). That density is close to the critical level that initiates a phase transition from solid to liquid. Therefore, it is reasonable to expect that the major contributions to the absorption are made by the free carriers and the inter-band transitions in all the three materials.

The fundamental mechanism of free-carrier absorption involves electron-phonon-photon collision [42]. To make an appreciable contribution to absorption, the electrons should experience a few collisions within the penetration depth [43-45]. Therefore, the free-carrier contribution to the absorption can be evaluated by comparing the penetration depth δ_n against free-electron mean free path l and the skin-layer depth δ_e for the skin effect exclusively produced by the free carriers [43-45]. The conduction-band electron mean free path is 2-6 times larger than the penetration depth in silicon (Table 1). In the titanium alloy, it is appreciably smaller than the penetration depth. Finally, the electron mean free path is very close to the penetration depth of stainless steel (Table 1). The exclusive free-carrier contribution to the normal skin-layer depth is estimated as follows [43, 44]:

$$\delta_e = \sqrt{\frac{2\rho}{\omega\mu_r\mu_0}} \quad (4)$$

where μ_r is relative magnetic permeability (that is about 1.0 for all the materials); μ_0 – magnetic permeability of vacuum; ω – laser frequency; ρ – resistivity of the material at laser frequency. Eq. (4) delivers approximately 8.5 nm depth for the titanium alloy and 15.5 nm for stainless steel. In silicon, the skin-layer depth of Eq. (4) exceeds 100 nm for any realistic conduction-electron density of the n-doped crystal.

Therefore, those estimations support the hypothesis about domination of the inter-band absorption at the laser wavelength in silicon and stainless steel, and domination of the free-carrier absorption in the titanium alloy. This conclusion is also supported by the fact that atoms of heavy metals, e.g. vanadium strongly contribute to the electron scattering in titanium alloys [31, 32]. The scattering significantly reduces the mean free electron path and increases the probability of electron-defect interactions that correlate with increased resistivity of titanium alloys [30-32].

The contribution of inter-band electron transitions to absorption of silicon can be significant because photon energy of laser radiation (3.5 eV) exceeds the direct bandgap of silicon at Γ and L points of the Brillouin zone (3.2 – 3.4 eV) and the indirect band gap (1.12 eV) [46]. The photon energy also exceeds the valence-to-conduction gap in iron [25, 26]. Those facts further support domination of the inter-band excitations over the free-carrier absorption in silicon and stainless steel at laser wavelength.

Since the dominating absorption mechanisms are different in the materials, it is required to analyze the magnetic-field influence on both the free-carrier and inter-band contributions to the absorption. The free-carrier interaction with the magnetic field can result in anomalous penetration phenomena [43, 44] that increase the depth of energy deposition in the magnetic field. However, they can hardly be significant in the metals because the magnetic field is not strong enough and the temperature is not low enough to meet the necessary requirement for the magnetic-field-induced penetration effects [43, 44]:

$$R_L < l \quad (5)$$

where l is mean free path for electrons, and R_L is a value of Larmor radius of the conduction electrons in the metals. The ratio R_L/l is of the order of 10^2 – 10^3 in the experiments. Also, the cyclotron frequency of the conduction electrons is of the order of 10^{10} – 10^{11} Hz that is 10^2 – 10^4 times smaller than the electron-particle collision rate in typical metals [30-32]. Therefore, the electron-particle collisions are frequent enough to destroy any magnetic-field-driven circular trajectory of the conduction electrons and prevent any remarkable influence of the magnetic field on the free-electron absorption in the two tested metals.

The influence of magnetic field on the inter-band absorption is attributed to driving a cyclic electron motion, splitting the original valence and conduction bands into Landau sub-bands, and driving resonant transitions between them [42, 47, 48]. First, we consider the inter-band electron excitation in the metals [45, 46]. As

estimated just above, the electron-particle collisions dominate over the magnetic-field-driven cyclic motion of electrons. Also, the quantizing energy of the magnetic field $\hbar\omega_c$ is substantially lower than the average thermal energy of the free electrons $k_B T$, and the thermal motion completely smears the Landau sub-bands [49]. Therefore, those processes destroy the Landau sub-bands, and the magnetic field can hardly produce any remarkable influence on the inter-band excitation in the metals to increase absorption and improve laser-surface coupling [42, 49].

The situation is different in silicon because the small effective mass of the conduction-band electrons [46] supports a higher cyclotron frequency in the range from 0.32 THz (Γ point of the conduction band) to 0.46 THz (L valley of the conduction band). On the other hand, the electron-particle collisions are substantially less frequent than in the metals due to the reduced electron density in the conduction band prior to the laser action. According to the simulations of the electron mean free path in silicon at room temperature [29], the electron-phonon collision time is estimated as 300-400 fs at zero magnetic field prior to the laser action assuming the average thermal speed of the electrons [46]. The associated collision frequency is 2.5 - 3.3 THz that is just 5.4 times larger than the cyclotron frequency in the ablation experiments. Also, the smallest Larmor radius of the conduction electrons is about 350 nm compared to 60 nm mean free path [29]. These data suggest the conduction electrons can make at least 1/6 of a full cyclotron cycle on average in the silicon crystal.

However, the magnetic field can appreciably reduce the rates of electron-electron and electron-phonon collisions [49, 50] even at moderate values of the magnetic field. The most significant reduction is for the rate of electron-electron collisions responsible for the fastest de-phasing of magnetic-field-driven cycling [50]. Among various electron-phonon collisions, electron collisions with polar phonons are the fastest [42]. Their rate can experience a few-fold reduction by the magnetic field [49, 50]. Those effects are highly supportive for extending of the partial cyclotron motion of the electrons and holes towards a full cyclotron cycle. Therefore, they can enhance the influence of the axial magnetic field on the laser-surface coupling and optical response. In particular, the increase of penetration depth and modification of surface reflectivity can be expected [43, 44]. The cyclotron motion leads to a more pronounced splitting of the initial energy bands into the Landau sub-bands and excitation of resonant transitions between them [23, 48]. More important, the axial alignment of the magnetic field leads to modification of selection rules and activation of the inter-band electron transitions that are forbidden without the magnetic field [46, 48]. Therefore, the magnetic field can appreciably increase the absorption of silicon and improve the laser-surface coupling. Those effects are significant enough to justify the 1.3-fold increase of the ablation rate by single-pulse ablation [23].

C. TRANSPORT AND THERMOMAGNETIC EFFECTS

The magnetic field can also affect another group of processes that make a crucial contribution to the laser

ablation – transport processes, e. g., transfer of heat. First, we note that a solid must be heated to at least its melting point to initiate some material removal by the thermal ablation mechanisms. Therefore, the very surface layer of the ablated materials is liquid with an interface between the melted layer and heated crystalline part of the solid (see Fig. 6). It is notable that the melting and boiling points of the titanium alloy are the highest compared to silicon and stainless steel (Table 2). This fact might explain the absence of an appreciable ablation of the titanium alloy. However, the heat-spreading distance (as estimated via thermal diffusivity of a solid phase at melting point – see Table 2) and absorption at laser wavelength (Table 1) have very similar values for stainless steel and titanium alloy (Table 2). Therefore, the linear scaling of peak temperature with laser fluence [51] suggests that increase of laser fluence by factor of two over the ablation threshold of stainless steel can lead to ablation of titanium alloy. Since that did not happen in the experiments (Fig. 5), other mechanisms were responsible for the observed ablation features.

Second, the high-temperature characteristic of the ablation suggests substantial temperature dependence of thermal conductivity λ_{TH} [52-53]. For some range of temperatures around a melting point, the thermal conductivity linearly scales with temperature [52-53]:

$$\lambda_{TH}(T) = \lambda_{TH0} + aT. \quad (6)$$

The most remarkable feature of the temperature dependence of thermal conductivity is that it makes a jump at the interface between the solid and liquid phases of silicon and stainless steel (Table 2; Fig. 6 (a) and (b)). In silicon, the thermal conductivity increases by approximately 100% in the liquid phase compared to that in the crystalline phase. In stainless steel, the thermal conductivity reduces by almost 50% in the liquid phase. In the titanium alloy, variations of thermal conductivity at the

liquid-solid interface are about 10%. Since the flow of thermal energy:

$$\vec{q} = \lambda_{TH}(T) \vec{\nabla} T, \quad (7)$$

is continuous at the liquid-solid interface [49], abrupt jumps of temperature gradients must be formed at the interfaces to compensate the step-wise changes of thermal conductivity. This implies variation of the temperature gradients by 50% within a nanometer-thick layer at the interfaces in silicon and stainless steel, but the variation is just by 10% in the titanium alloy (Fig. 6(c)).

The temperature gradients produce thermo-mechanical stresses [57, 58] that stimulate mechanical motion of the melted layer and support material removal attributed to the ablation. Therefore, formation of the large temperature gradients at the liquid-solid interfaces is highly supportive for ablation of silicon and steel, but the thermomechanical forces are substantially smaller in the titanium alloy. This qualitative conclusion perfectly correlates with the remarkable similarity of the ablation features on silicon and steel, and the specific behavior of the titanium alloy. The specific conditions at the liquid-solid interfaces are highly favorable for enhanced thermomagnetic effects that result from a modification of thermal conductivity by the magnetic field [49, 50, 54-56]. In general case, thermal conductivity contains contributions from free carriers and phonons [42], but the free-carrier contribution is the only one affected by the magnetic field [49, 50, 55]. In metals, the free-carrier thermal conductivity dominates [44], and the perturbation theory of the thermomagnetic effects [55-57] delivers the following linear scaling with the magnetic field:

$$\lambda_{TH}(T, \mathbf{B}) = \lambda_{TH}(T) + \sigma \mathbf{B} \cdot \nabla T, \quad (8)$$

where σ is a tensor in general case. The traditional models [55, 56] suggest that the magnetic fields employed in the reported above experiments produce negligible variations of thermal conductivity at room temperature.

Table 2. Thermophysical properties of silicon [52], stainless steel 316L [53], and titanium alloy Ti6Al4V [54].

Material	Boiling point [°C]	Melting point [K]	Thermal conductivity; solid at melting point [W/(m K)]	Thermal conductivity; liquid at melting point [W/(m K)]	Thermal diffusivity; solid at the melting point [cm²/s]	Thermal diffusivity; liquid at the melting point [cm²/s]	Density (solid) at 298 K [g/cm³]	Heat spreading distance within 20 ns (solid) [nm]	Heat spreading distance within 10 ns (liquid) [nm]
Si (crystal)	2653	1685	27.3	58.6	0.114	0.223	2.329	477	668
Stainless steel	2870 (Fe)	1700	35.96	17.98	0.0722	0.0335	8.00	380	259
Titanium alloy Ti6Al4V	3290 (Ti)	1900	26.69	29.03	0.074	0.058	4.42	385	341

Fig. 6. A sketch of distribution of temperature-dependent thermal conductivity (red solid) and thermal flow (blue dotted) in the melted surface layer and at the liquid-solid interface of silicon (a), stainless steel (b), and titanium alloy (c). Shaded strips depict location of high temperature gradients. Vertical axis sketches scale of thermal-conductivity variations in arbitrary units.

In silicon prior to laser action, the effect of magnetic field on thermal conductivity is negligible because the phonon mechanism of heat transfer dominates at low density of free carriers [42, 49, 50, 55]. However, the laser-induced generation of the free electrons due to the one-photon inter-band transitions rapidly increases the free-carrier density at the very beginning of a laser pulse. Therefore, the free-carrier contribution to thermal conductivity can dominate over the phonon one over a major part of the laser pulse. In the magnetic field, the electron excitation results in dynamic enhancement of the thermomagnetic effects in silicon. More detailed analysis of the thermomagnetic effects at the heated liquid-solid interface meets some fundamental challenges. The perturbation-type theories of the effects [49, 5, 55, 56] assume small values of temperature gradients and consider the linear scaling of Eq. (8) with both magnetic field and temperature gradient. That scaling may be violated at the liquid-solid interfaces in silicon and stainless steel because the large temperature gradients may produce nonlinear and anomalously strong effects. Therefore, nonlinear models of the thermomagnetic processes are required for analysis of the magnetic-field-enhanced ablation. Such models are not available for the materials under consideration.

D. OTHER MAGNETIC-FIELD EFFECTS

The magnetic field can also influence the process of material removal during ablation. First, it can affect the early stage of LPP formation by magnetization of the atomic clusters that depart from the surface. Analysis of this effect requires extended simulations of the material transfer and early stages of LPP dynamics in the axial magnetic field that will be reported elsewhere.

Second, the axial magnetic field can affect the thermomechanical stresses at the liquid-solid interface. A straightforward mechanism of this influence is attributed to variations of the interface temperature gradients by the thermomagnetic effects [54, 55]. For relatively low temperature gradients, the thermomechanical stresses scale linearly with absolute value of the gradient [58, 59]. However, the possible large values of temperature gradients localized at the interfaces are favorable for nonlinear thermomagnetic effects, e. g., a nonlinear analog of the Righi-Leduc effect [55] and may lead to a nonlinear scaling of the stresses with temperature gradients. Those effects can produce extra-normal and tangential

temperature gradients at the interfaces. Detailed analysis of those processes requires extended simulations that are beyond the scope of this paper.

Third, the magnetic field can affect hydrodynamic motion of the melted materials [59]. The physical effects that support the influence of magnetic field on the melt flow are attributed to the presence of ions in the melt. Therefore, the hydrodynamic flows carry some charges and produce the electric currents influenced by the magnetic field. Simulations of the hydrodynamic effects of magnetic-field-assisted laser welding [59] suggest feasibility of a significant modification of the melt flows by moderate magnetic fields. The magneto-hydrodynamic effects are likely to contribute to the magnetic-field-induced modification of morphology of the melt rims at the groove edges (Figs. 3, 4, and 5). However, detailed analysis of those processes requires extended simulations and is a subject of a separate research to be reported elsewhere.

Finally, the specific morphology of the laser-modified roughness of the titanium-alloy surface (Fig. 5) suggests a significant oxidation of the laser-treated area. The oxidation may happen in the plasma plume [60] followed by back deposition of the oxides. Also, a direct oxidation of the heated surface may take place. The substantial variation of the surface morphology of the titanium alloy by the axial magnetic field points at a significant influence of the magnetic field on the oxidation process (Fig. 5(b)). Detailed studies of those processes are beyond the scope of this paper.

5. CONCLUSION

Grooves were ablated on silicon, stainless steel (316L), and titanium alloy (Ti6Al4V) under the axial magnetic field. The results showed a significant similarity between silicon and stainless-steel: the magnetic field increased the groove depth by factor of 2.8 – 3.0, reduced groove width, and suppressed back-deposition of large ablated pieces. The titanium alloy behaved in a qualitatively different way and showed specific surface modification by the increase of roughness at no pronounced material removal. The axial magnetic field modified morphology of the roughness but did not stimulate removal of the titanium alloy.

Analysis of the fundamental mechanisms of the magnetic-field-induced ablation enhancement was focused on identification of the physical effects that can drive the specific ablation behavior of those materials. A

very significant correlation was found between the major ablation features and the formation of temperature gradients at the solid-liquid interfaces due to the step-wise variation of thermal conductivity. Large values of the gradients are associated with the pronounced ablation of silicon and stainless-steel surfaces while very minor gradients are attributed to the weak ablation of the titanium alloy. The large temperature gradients localized at the solid-liquid interfaces can nonlinearly enhance various thermos-magnetic effects and contribute to both heat transfer and further generation of the thermomechanical stresses at the interfaces. A detailed analysis of those effects requires systematic and extended simulation efforts that go far beyond the scope of this paper. Other physical effects that can appreciably contribute to the reported magnetic-field-induced enhancement of ablation include (a) modification of plasma properties and composition by the magnetic field; and (b) magnetic-field-induced enhancement of absorption in silicon. Some contributions are expected from a magnetic-field influence on hydrodynamic motion of melt and of oxidation of the metals. We are considering further experimental efforts focused on more specific identification of the contributions of those fundamental mechanisms to the ablation enhancement under the axial magnetic field.

From the viewpoint of potential applications, the axial magnetic field provides a unique capability to fix the fundamental issues of the nanosecond laser-surface coupling. This approach has shown a significant increase of the ablation depth, reduction of ablation threshold, and reduced back-deposition of ablation debris. It is more effective when applied to semiconductors (e.g. silicon) compared to metals (e.g. stainless-steel and titanium alloy). Proper use of the axial magnetic field opens a new pathway to more efficient, faster, and better-quality laser micro- and nano-machining with superior potential for new approaches in laser additive manufacturing.

Acknowledgment. The authors acknowledge A*Star SINGA scholarship and Singapore Institute of Manufacturing Technology (SIMTech) for their support. Dr. Wolfgang Rudolph of the University of New Mexico is acknowledged for a kind suggestion to consider the skin-layer effects.

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