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Magneto-absorption effects in magnetic-field assisted laser ablation of silicon by UV nanosecond pulses

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A constant magnetic field can significantly improve the quality and speed of ablation by nanosecond laser pulses. These improvements are usually attributed to the confinement of laser-produced plasma by the magnetic field and specific propagation effects in the magnetized plasma. Here we report a strong influence of constant axial magnetic field on the ablation of silicon by 20-ns laser pulses at wavelength 355 nm, which results in an increase of ablation depth by a factor of 1.3 to 69 depending on laser parameters and magnitude of the magnetic field. The traditional plasma effects do not explain this result, and magneto-absorption of silicon is proposed as one of the major mechanisms of the significant enhancement of ablation. *Published by AIP Publishing.*

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Improved coupling between laser-pulse energy and solid surfaces is among the major challenges for optimization of multiple high-power laser-surface interactions, e.g., micro-machining by laser ablation, laser-induced breakdown spectroscopy, pulsed-laser deposition of thin films, and laser-driven chemical reactions.^{1–21} Laser-produced plasma (LPP) is frequently assumed to be the major effect that distorts laser-surface coupling and shields the target surface from the incident laser beam.^{1–8} Based on the concept of LPP confinement by moderate magnetic fields of 0.1 to 1.0 T,^{14–21} application of a constant magnetic field during laser-surface interactions^{9–13} is considered as an effective approach to address that issue. However, almost all previously reported experiments on magnetic-field-assisted ablation and LPP interactions were performed with conducting surfaces,^{9–21} the majority of which were paramagnetic metals (aluminum, copper, and tin). Because of the specific magnetic properties of those metals, modifications of material response by the magnetic field were completely ignored.^{9–21} Also, the influence of magnetic field on ablation of non-metal surfaces has received very minor attention. In this study, we find a very strong influence of an axial magnetic field on the ablation of silicon by UV nanosecond laser pulses that increases single-pulse ablation rate by a factor of 1.3. For multipulse ablation, the rate increased by a factor of 4 to 69 depending on irradiation parameters and the magnitude of the magnetic field. The traditional concept of ablation affected by magnetized LPP cannot fully explain our observations. Therefore, we consider the possible influence of magneto-absorption of silicon to account for the significant enhancement of ablation.

The surface used in the experiments was a (100) plane of a single crystal n-type (phosphorous doped) silicon wafer with a thickness of 300 μm . The wafer was cut into $10 \times 10 \text{ mm}^2$ pieces. Ultrasonic cleaning of the samples was carried out in methanol for 10 min, in acetone for another 10 min, and then in de-ionized water for 30 min. A Q-switched third-harmonic Nd:YVO₄ laser delivered 20-ns pulses with a Gaussian beam profile at wavelength 355 nm and repetition rate 1 kHz to conduct the experiments in air. The mean laser spot diameter was 50.94 μm (at 50% of maximum of intensity; averaged over 5 measurements). For generating a uniform axial magnetic field of 0.05–0.4 T, we used a copper coil with a current passing through it. The direction of magnetic field was reversed by changing poles of DC power supply. A silicon sample was mounted deep inside the coil to provide a uniform magnetic field during ablation. Characterization of ablated craters was done by scanning electron (SEM) and confocal optical microscopes.

Typical depth profiles of ablation craters and their SEM images are shown in Fig. 1. The silicon wafer was irradiated at a laser-pulse energy of 112 μJ by applying either 1 or 10 pulses per single site. A single pulse produced ablation depth of 3 μm without a magnetic field (Fig. 1(a)). SEM (Fig. 1(a)) and optical microscope image (Fig. 2(b)) confirmed significant deposition of ablation residuals around the crater rim. Ablation with the same irradiation parameters under a magnetic field of 0.192 T directed toward the surface led to an increase of crater depth to 4 μm (Fig. 1(c)) and reduction of crater diameter from 40 μm (no magnetic field) to 25 μm . For 10 pulses, the depth increased from 10 to 37 μm (Fig. 1), and the reduction of the crater diameter significantly improved aspect ratio. Furthermore, the magnetic field reduced the size of debris particles around the crater (Figs. 1 and 2), but increased the area of debris deposition. Slightly asymmetric

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FIG. 1. Representative profile of ablation crater produced by 1 (a) and (c), and by 10 (b) and (d) laser pulses at a pulse energy of $112 \mu\text{J}$ and a laser fluence of 3.81 J/cm^2 . Magnetic field is either zero (a) and (b) or 0.192 T (c) and (d). Direction of magnetic field is shown by blue arrows; inserts are SEM images of ablation craters.

distribution of debris was attributed to minor misalignment and asymmetry of the laser beam. Dependence of ablation depth on laser-pulse energy (Fig. 2(a)) demonstrated strong reduction of the ablation threshold by the magnetic field for both single- and multi-pulse ablation.

For interpretation of those results, feasibility of LPP confinement by magnetic field is checked by evaluation of cyclotron frequency and cyclotron period following the traditional concept.^{9–21}

$$\omega_C = \frac{qB}{m}; \quad \tau_C = \frac{2\pi}{\omega_C}, \quad (1)$$

where m is mass of a charged plasma particle; q is the particle charge; and B is magnetic field; all values are in SI units. For heavy-mass component of LPP, the shortest cyclotron period of $9.54 \mu\text{s}$ is attributed to a single-charged ion of Si

($m = 28.085$ atomic units of mass;²² $\omega_C = 0.659 \times 10^6 \text{ rad/s}$ at $B = 0.192 \text{ T}$). Within a pulse duration of 20 ns , that particle makes a very negligible angular travel of 0.013 rad , i.e., practically does not deviate from a trajectory made without a magnetic field. Therefore, significant distortions of dynamics of heavy LPP particles by magnetic field are delayed by few hundreds of nanoseconds in agreement with previously reported delays of the magnetic-field driven evolution of LPP.^{10–21} Therefore, the heavy-mass LPP component is not confined by the magnetic field during direct laser action and early stages of ablation, but major confinement effects occur at the stage of plasma cooling and deposition.

More insight into the interactions of the laser beam with magnetized LPP is provided by the Larmor radius, which can be estimated by assuming that the initial speed of plasma particles departing from the surface is the same as the average thermal speed of silicon atoms in liquid phase at boiling point T_{BOIL} :

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

where k_B is the Boltzmann constant; $\bar{v}$ is average particle speed. For $T_{\text{BOIL}} = 3558 \text{ K}$,²² the average speed is $1.77 \times 10^3 \text{ m/s}$, and the Larmor radius is 2.69 mm for a single charged silicon ion. During ballistic expansion, the speed of the plasma front can reach 10^5 m/s ,^{14–21} which increases the Larmor radius by two orders of magnitude. This result explains the specific distribution of debris around the craters (Figs. 2(b) and 2(c)) since massive and energetic LPP particles tend to move further from the crater at the stage of plasma cooling and debris deposition according to Eq. (2). However, those delayed effects can hardly affect the depth of the ablation crater.

Propagation effects in magnetized LPP are the more probable cause of enhanced ablation. They are controlled by the optical response of LPP given by the Appleton formula for propagation direction parallel to magnetic field:²³

FIG. 2. (a) Ablation-crater depth vs laser-pulse energy for zero-magnetic field (blue) and 0.192 T magnetic field (red) done with 1 pulse (dashed) and with 10 pulses (solid). The vertical error bars obtained from 5 measurements for each of the four values of laser-pulse energy (16 , 35 , 56 , and $112 \mu\text{J}$) do not exceed the size of the markers in most cases. Optical microscope image depicts ablation craters produced by 1 pulse with zero (b) and 0.192 T magnetic field (c).

$$\epsilon_{LPP} = 1 - \frac{(\omega_p/\omega)^2}{1 - i\nu_e/\omega - \omega_c/\omega}, \quad (3)$$

where ω is laser frequency, ω_p is plasma frequency, and ν_e is the effective collision frequency. The low value of cyclotron frequency for heavy LPP particles (ω_c/ω is about 10^{-10} – 10^{-9}) implies their negligible contribution to the modification of the LPP optical response in the presence of a magnetic field. Cyclotron frequency of LPP electrons is 3.377×10^{10} rad/s at $B = 0.192$ T, but the ratio ω_c/ω is still about 10^{-5} . In spite of the negligible modification of LPP dielectric function by the magnetic field, some specific propagation effects still can affect the delivery of laser energy through the magnetized plasma.²³ Their contribution can be easily checked because propagation through a magnetized plasma is sensitive to mutual alignment of the magnetic field vector and the wave vector of the laser beam.²³ Bearing this fact in mind, we produced three grooves on the same sample at a laser-pulse energy of 35 μ J (Fig. 3), which was close to the ablation threshold (threshold fluence 1.17 J/cm²; threshold irradiance 55.15 MW/cm²) with the given laser parameters.^{24,25} One of the grooves was made under no magnetic field (depth 0.142 μ m, Fig. 3(a)), another under 0.341 T magnetic field directed toward the surface (depth 9.76 μ m, Fig. 3(b)), and the third one under 0.341 T magnetic field directed away from the surface (depth 9.75 μ m, Fig. 3(c)). The LPP propagation was the only effect expected to be sensitive to alignment of the magnetic field, and the experimental data suggested those effects did not significantly influence ablation depth. However, the reduction of crater diameter (Fig. 1) suggests that some plasma-propagation effects contribute to the obtained results and can be associated with electrons in the plasma confined within the central part of LPP due to high cyclotron frequency (6.0×10^{10} rad/s) and the smaller Larmor radius of electrons (18–33 μ m for average electron energy equal to one laser photon).

Other effects produced by interactions of the magnetic field with LPP are the self-focusing of the laser beam by non-homogeneous magnetized plasma²⁶ and the heating of the surface by thermal radiation of plasma.²⁷ Self-focusing is predicted by simulations for magnetic fields parallel to propagation direction,²⁶ but very high magnetic fields²⁶ ($\omega_c/\omega = 0.1$ – 0.5) are required to make the effect appreciable for considered LPP extension (2–5 mm). This effect remarkably contributes only for multi-pulse ablation of high-aspect-ratio craters, since confinement of LPP by a deep crater can produce significant density gradient to support the self-focusing.²⁶ The thermal radiation of plasma can contribute 10%–20% to the increase of ablation depth following the simulations for graphite,²⁷ but this contribution does not account for the huge increase in ablation depth.

Finally, the ablation enhancement can result from modification of the optical properties of silicon by a magnetic field.^{28,29} The N-type of ablated silicon is favorable for a contribution of free conduction electrons to absorption modification²⁹ attributed to the formation of two minima on the reflectivity spectrum. The minima are symmetrically shifted from the unperturbed electron-hole plasma frequency ω_p (Fig. 4(a)). Effective masses of electrons and holes of silicon vary from 0.14 to 0.9 of free-electron mass,²² and corresponding cyclotron frequency varies from 3×10^{11} down to 6×10^{10} rad/s in the utilized magnetic fields. However, all the reflectivity minima are in mid-infrared,^{28,29} and the magnetic-field interaction with free carriers must not affect reflectivity and absorption at the laser wavelength.

Absorption of semiconductors is also modified by splitting of original energy bands into Landau levels and activation of resonant transitions between them^{28–30} (Fig. 4(b)). That effect results in the formation of multiple maxima near the edge of the absorption band formed by direct inter-band transitions²² at 3.2–3.4 eV (Fig. 4(c); unperturbed absorption

FIG. 3. Profile of ablation craters produced by linear laser scanning with speed of 1 mm/sec, pulse energy of 35 μ J, and fluence of 1.19 J/cm² with no magnetic field (a) and (d); under magnetic field 0.341 T directed toward the surface (b) and (e); and under magnetic field 0.341 T directed away from the surface (c) and (f). 3D images of the ablated grooves (d)–(f) shown under corresponding profiles are acquired by the confocal optical microscope.

FIG. 4. (a) A sketch of spectrum of silicon reflectivity due to electron-hole plasma with zero (blue) and non-zero magnetic field (red). (b) Band-structure diagram of silicon with possible transitions with no magnetic field (blue arrow) and in the magnetic field (red arrows). Green areas depict location of the Landau levels involved into the direct inter-band transitions in the magnetic field. (c) A sketch of the edge of absorption spectrum attributed to the direct inter-band transitions under zero (blue line) and non-zero magnetic field (red line).

spectrum is obtained from tables of Ref. 31; modification of the spectrum by magneto-absorption for inter-band transitions are given by equations of Ref. 29 for magnetic field orthogonal to electric field of laser light). Small values of cyclotron resonance for free holes and electrons of silicon result in extra-small energy gaps between the Landau levels that merge and form quasi-continuums favorable for resonant inter-band transitions at the laser wavelength. Those transitions produce resonant increase of absorption of laser light in the magnetic field. This effect can be substantially enhanced by extra inter-band transitions resulting from modification of selection rules by the magnetic field.^{30,31} In particular, our experiments employ orthogonal alignment of the constant magnetic field and the electric field of laser radiation that unlocks inter-band transitions between heavy-hole and light-hole valence bands^{28,29} (Fig. 4(b)). The rate of those transitions scales as $(\hbar\omega - \epsilon_{\text{eff}}(B))^{-1/2}$ with energy of laser photons and the magnetic field,²⁹ where $\epsilon_{\text{eff}}(B)$ is energy gap between Landau levels of the two valence bands involved into the transitions. Therefore, the orthogonal alignment of laser polarization and the magnetic field is especially favorable for a resonant increase of absorption due to the magneto-absorption effects. This increase can be very significant if the energy of laser photon fits the energy gap between the quantum states involved into the resonant transitions (Figs. 4(b) and 4(c)). Detailed analysis of the influence of magneto-absorption effects on ablation deserves a separate study and will be reported elsewhere.

In summary, the significant increase of crater depth and aspect ratio, and reduction of the ablation threshold are produced by magnetic fields applied during ablation of n-type silicon by UV nanosecond laser pulses. Qualitative analysis shows that modification of propagation conditions of laser pulses in magnetized LPP and enhancement of the absorption of silicon by a magnetic field can be the most effective causes of these changes to laser ablation. Demonstrated ablation enhancement can significantly alter the present view of laser ablation of semiconductors and related manufacturing technologies. It opens a pathway to making them more efficient and faster without the consumption of extra power by industrial lasers.

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