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Measuring single-shot, picosecond optical damage threshold in Ge, Si, and sapphire with a 5.1-µm laser

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Abstract: Optical photonic structures driven by picosecond, GW-class lasers are emerging as promising novel sources of electron beams and high quality X-rays. Due to quadratic dependence on wavelength of the laser ponderomotive potential, the performance of such sources scales very favorably towards longer drive laser wavelengths. However, to take full advantage of photonic structures at mid-IR spectral region, it is important to determine optical breakdown limits of common optical materials. To this end, an experimental study was carried out at a wavelength of 5 µm, using a frequency-doubled CO2 laser source, with 5 ps pulse length. Single-shot optical breakdowns were detected and characterized at different laser intensities, and damage threshold values of 0.2, 0.3, and 7.0 J/cm², were established for Ge, Si, and sapphire, respectively. The measured damage threshold values were stable and repeatable within individual data sets, and across varying experimental conditions.

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The experimental determination of damage thresholds of optical wafers, reported herein, was motivated by the development of the dielectric laser accelerator (DLA), a promising laser-powered technology to reduce the characteristic feature sizes of electron beam sources down to the optical scale [1]. DLAs can reach accelerating gradients exceeding 1 GeV/m, which is more than an order of magnitude improvement over the state-of-the-art radio frequency linear accelerators (RF linacs) presently dominating research and industrial applications. The DLA has been considered for decades [2-4], but only recently has the concept gained momentum [5,6] with advances in solid-state laser technology and nanoengineering methods. A significant milestone—the first experimental demonstration of DLA energy gain—has been achieved at the Stanford Linear Accelerator Center (SLAC) in 2013 [7].

Most of the DLA work to date has been done at 800 nm due to the commercial availability of Ti:Sa lasers. A number of DLA topographies are being considered [8,9], including photonic...
band gap fiber and crystal structures [10,11] and transmission-mode grating structures [12]. To relax the spatial acceptance limitations in at least one dimension, planar standing-wave (SW) DLA structures have been developed [13]. Nevertheless, at least one dimension of the electron beam’s transverse size has to be confined to a fraction of the laser wavelength, generating strong interest in scaling DLA structures to mid-IR. Recently a new traveling-wave photonic structure DLA was proposed at UCLA [14], operating at 5 µm to take advantage of the emerging availability of high-power, mid-IR, OPA sources [15]. While the 5-µm laser technology itself is more challenging, it is a rapidly evolving field that is also driven by the demands of high-harmonic generation (HHG) [16,17]. Thus, it is expected that mid-IR laser sources suitable for driving DLAs will be commercially available within a few years.

The use of longer wavelength radiation offers several advantages that enable practical DLA devices. Expanding the gap size permits a relaxation of all structure dimensions and fabrication tolerances. Larger structure apertures and stored energies allow higher charges to be accelerated, mitigating both longitudinal and transverse wake-field effects and permitting larger source emittances. Finally, the larger achievable beam loading plays an important role in improving the overall energy efficiency of the acceleration system—a critical consideration for DLAs in the context of future linear collider technology [18].

One important factor, however, is the picosecond optical breakdown limit at mid-IR wavelengths. In general, for any resonant accelerating structure, the maximum achievable gradient is always limited by a breakdown effect. A typical industrially available S-band (3-GHz) microwave linac usually operates at a gradient of about 20 MV/m, and some high performance linacs designed for collider applications can reach up to 100 MV/m at 11 GHz [19]. Beyond these values, however, RF breakdown events are unavoidable and constitute a fundamental limitation in the practical use of microwave technology at high gradients. Since the breakdown threshold increases with operating frequency and is higher for dielectrics [20], it is believed that a DLA will be able to sustain field gradients exceeding 1 GV/m with picosecond pulses. However, before such a DLA photonic structure can be implemented in practice, a picosecond laser damage threshold needs to be established for common photonic building materials.

The general principles for the damage mechanisms near threshold from short-pulse (picoseconds or shorter) lasers are well understood [21-22], and the theory is well supported by experimental data in the visible and near infrared (NIR) spectral ranges [23-26] including some recent experimental studies which were conducted at SLAC [27] in the context of the DLA development. There is, however, no prior data on the short-pulse breakdown in the mid-IR range. Thus, to facilitate development of mid-IR DLA structures, we investigated the breakdown threshold of three materials: Ge and Si and sapphire (Al₃O₃). We have chosen the popular semiconductors Ge and Si for manufacturability. The fabrication processes for sapphire is less established, however the material has a much larger band gap than either germanium or silicon, and all the experimental data in the optical spectral range [23-27] indicated an anticipated breakdown threshold dependence on the band gap. Thus, sapphire was chosen as an important reference point for comparing mid-IR to NIR optical breakdown properties.

2. Experiment

The experiments were performed at Brookhaven National Laboratories’ Accelerator Test Facility (ATF). The short-pulse CO₂ laser design and operation have been described in detail elsewhere [28]. At ATF, the 5-ps CO₂ pulses of variable peak power—up to TW levels—are available. The 10.2-µm pulse is frequency-doubled to 5.1 µm using ZnGeP₂ (ZGP) non-linear crystal, which at longer wavelengths demonstrated superior performance to other doubling crystals [29] in having the highest nonlinear coefficient and the highest damage threshold.
Initial experiments were conducted in air, with the system set on an optical table, as shown in the photograph in Fig. 1. The CO2 beam initially used was directly from the regenerative amplifier, generating 5 ps pulses at 10.2 µm with the pulsed energy up to 10 mJ. However, at the location of ZGP crystal (in a different room) the transport resulted in losses of about 55% of the initial CO2 beam power, resulting in up to 4.5 mJ delivered for second harmonic generation (SHG). Shot-to-shot energy fluctuated over a wide range (2.0–4.5 mJ), but the spatial profile of the laser was very stable and clean (M² ~ 1.6) with the waist size on the order of 200 µm.

Fig. 1. A photograph of the in-air system on an optical table, and a schematic diagram of the in vacuum set up. A 10.2-µm laser pulse is matched into ZGP SHG crystal, and generates a 5.1-µm beam, which is separated from the 1st harmonic background via a set of dichroic mirrors, and then focused on a sample. A joulemeter and a pyro camera enable the tracking of sample exposure for every shot.

SHG was optimized in two steps: (1) the crystal was rotated in the transverse and horizontal planes to determine the angle where the process was most efficient to accommodate for the small difference between the crystal design wavelength (10.6 µm) and CO2 laser operating wavelength (10.2 µm), (2) the crystal longitudinal position with respect to the CO2 laser waist was scanned and optimize to avoid crystal damage and saturation. The optimized position was determined at about 4 cm upstream of the waist, where the CO2 spot size was about 450 µm, and at 4.5 mJ the conversion efficiency of 14% was achieved, yielding 600-µJ pulses at mid-IR. In the later round of measurements (for the sapphire samples), the CO2 pulse was passed through an additional amplification stage to improve stability and achieve higher pulsed energy levels at the SHG crystal, but the optimization procedure was the same.

A picosecond, 5.1-µm, second-harmonic pulse generated by SHG is then separated from the background 10-µm light via a set of dichroic CaF2 mirrors and focused on the surface of a sample. A portion of the pulse is directed to the pyro detector, which provides shot-to-shot pulsed energy information. After the sample chamber there is also a pyro camera, which provides shot-to-shot images of the mid-IR beam. The IR camera is used to monitor the beam profile throughout the measurements as well as for initial alignment of the 5-µm beam to the reference pinholes positioned at the sample plane. An absolute measurement of mid-IR transmission through calibrated reference set of pinholes is used to derive the peak intensity at the laser plane. The pinholes also aid in the alignment of the in-situ damage detection camera.
A sample holder enabled transverse translation of the sample, so a new area of the sample is exposed after each consecutive shot. For each shot, the pulsed energy was imaged and the location was recorded and stored for comparison with the forensic analysis upon removal of the sample. Alignment and focal properties of the laser pulse were periodically sampled and remained consistent throughout the entire procedure. After the initial round of experiments, the vacuum chamber was introduced around the sample area, to make sure that an exposure to air does not affect the measured breakdown threshold.

3. Results

The samples were Si and Ge (001) single crystal wafers. The sapphire crystal was oriented along the <0001> direction. Both Ge and Si wafers were N-type with 53.3 to 57.9 Ω-cm resistivity. The samples received exposure from 5.1-µm laser pulses with 0.1–1-mJ energy, 60-µm diameter, and about 5-ps pulse length. After each consecutive shot the samples under test were shifted transversely by about 2 mm in a matrix pattern, and for each shot the matrix position on the wafer and the laser pulse intensity were recorded. The surface damage pattern was first detected in real time with the high magnification CCD in the measurement setup, and later examined forensically by scanning electron microscopy (SEM) using a Hitachi 4800 SEM in the secondary electron imaging mode.

Figure 2 shows SEM images of Ge (top) and sapphire (bottom) damage sites. The damage sites in all tested materials show both melting and ablation. In Ge, the large circular transition from dark to light indicates the limit of the melt zone, and the light ring is the frozen surface wave produced by the ablation event. In sapphire, more complex damage morphology is observed where the melt zone starts further away from the center, but following the same concentric surface wave pattern, often with multiple rings (very similar to the model introduced in [30]). In all samples (Ge, Si, and sapphire) there is a visible accumulation of larger ablation debris at the center of the laser damage trace.

The size of the damaged area was determined digitally measuring the radius of the ablation area. The peak fluence leading to the damage was determined by approximating the ablation area diameter to zero, whereas diameter of the ablation area was correlated with the laser
power, as shown in Fig. 3. The shot-to-shot laser intensity variations necessary for such measurement were naturally present, due to the CO₂ laser pulsed energy fluctuations (originating in the regenerative amplifier). For each set of measurements, the peak fluence threshold was obtained as a fit parameter to the curve in Fig. 3 (right). The beam width shot-to-shot variations were insignificant.

The measurements results are summarized in Table 1. For each set of data, the peak fluence resulting in damage to the optical surface was determined using the method shown in Fig. 3. The laser damage threshold is defined as a total pulsed energy in 1/e² laser beam (as per guidelines of ISO 11254). Thus, in a Gaussian beam approximation, the laser damage threshold value is simply ½ of the measured peak fluence value.

The measurements were first carried out in air, and later repeated in vacuum, using a specially fabricated measurement chamber to enable UHV level better than 10⁻⁵ torr. All samples were cleaned prior to shipment and handled in sealed enclosures until installation to remove particulate contamination from the surface. However, the in-air measurements were conducted without any clean room environment provisions, thus some reasonable degree of surface contamination was anticipated. Nevertheless, no quantitative difference in the damage threshold was observed for in-vacuum and out-of-vacuum measurements. Qualitatively, there were also no observable differences in the shape or pattern of the damaged area.

4. Discussion

Ge and Si are covalently bounded semiconductors [31] with similar electronic structures. They have indirect band gaps of 0.67 eV and 1.1 eV oriented along the <100> direction. Both materials have excitons bound just below the conduction band (at 0.004 eV and 0.015 eV), which is considered to be too close to significantly impact electron seeding. By comparison, sapphire has a strongly ionic character [32] and a direct band gap of 9.9 eV. Its excitons are
bound 1.1 eV (ordinary ray) and 1.2 eV (extraordinary ray) below the conduction band. So the optical band gap at ~8.8 eV provides a secondary ionization channel requiring fewer photons (or a lower electric field).

The general principles for picosecond laser-induced damage near threshold are summarized in [22] by Schaffer et al. Electrons are excited into the conduction band either through multiphoton absorption or tunneling ionization. In addition, while in the conduction band, free electrons can be further accelerated by the laser field, seeding the sequential ionization processes. Such avalanche ionization is an exponential process, and for longer pulses accounts for most of the absorbed laser energy. Regardless of the ionization mechanism, the electrons transfer their thermal energy to the lattice on a time scale on the order of 10 ps, and then they relax back to the valence band over a longer time determined by the carrier lifetime (typically microseconds).

An important insight into the damage mechanism can be drawn from the data statistics and also by comparing particulate-free in-vacuum measurements to those made in open air. For the short laser pulses (a few ps or less), the damage dynamics is significantly different then for the longer (> 10 ps) pulses [22]. In the long pulse regime, the exponential avalanche ionization process is the dominant mechanism, and its seeding is generally strongly influenced by surface defects. Thus, it is expected that the long-pulse, single-shot damage statistics have a strong dependence on the surface and experimental conditions and exhibit large shot-to-shot fluctuations. Quite to the contrary, the single-shot damage threshold measurements reported herein indicate relatively narrow (< 10%) variation in the breakdown limit throughout the entire data set and insensitivity to the surface and vacuum conditions. This supports an understanding that the observed surface damage was dominated by direct ionization mechanisms (i.e. tunneling) and not by the surface defects, vacuum conditions, and contamination. Thus, the interaction with the optical surface with an intense mid-IR laser pulse of as long as 5 ps in duration can still – within the context of single-shot optical breakdown dynamics – be classified as within the short pulse regime.

The relative contributions of multiphoton and tunneling ionization depends on the Keldysh parameter [33], \( \gamma = \omega / e \sqrt{m c \varepsilon_0 E_g / I} \), where \( \omega \) is the angular frequency of the laser, \( e \) is the electron charge, \( m \) is the reduced effective mass, \( c \) is the speed of light, \( \varepsilon_0 \) is permittivity of free space, \( E_g \) is the band gap, and \( I \) is the laser intensity. For \( \gamma > 1 \), the multi-photon absorption provides the initial electron density. If \( \gamma < 1 \), tunneling dominates. In all cases, due to the relatively long wavelength, the damage threshold occurred at \( \gamma \ll 1 \), thus indicating that multi-photon ionization mechanism was suppressed in all three tested materials. This may explain the approximately 40% increase in the absolute measured breakdown values with respect to the near infrared results reported in [27] by Soong et al.; namely, 0.14 J/cm² and 4.9 J/cm² for silicon and sapphire, respectively, at 800 nm laser wavelength and with sub-ps laser pulse lengths. These numbers are also consistent with Cowan’s results [34], reporting a Si damage threshold of 0.2 J/cm² at 0.7 ps and 2.2 µm wavelength.

For DLA photonic structures, the maximum accelerator gradient is related to the damage threshold by \( E_p \approx \sqrt{F_{th}} / \tau c \varepsilon_0 \), where \( E_p \) is the peak accelerating field, \( F_{th} \) is the damage threshold, \( \tau \) is the laser pulse length. The measured optical damage in Si (see Table 1) corresponds to a maximum accelerating field of only ~ 400 MV/m for 5 ps pulses; however, for sub-ps pulses, this value can exceed 1 GV/m, validating the development of DLA structures with large group velocities [14]. Although sapphire is a much more difficult material for photonic structures manufacturing, in a sub-ps DLA configuration it can support accelerating gradients exceeding 5 GV/m.
5. Conclusions

In summary, we report the first measurements of the single shot laser damage thresholds of Ge, Si, and sapphire with a picosecond, mid-IR laser operating at 5.1-μm wavelength. The results indicate characteristic dependence on the materials’ band gap values and an approximate 40% increase in damage thresholds compared to the near infrared data. This increase is attributed to the strong suppression of multi-photon absorption cross-section at longer wavelengths. An additional indirect observation from the damage statistical data is that at mid-IR spectral region, laser pulses of ~5 ps were insufficiently long to allow for avalanche ionization to play a significant role.

The initial motivation for this work was to assist the design of a high-gradient dielectric laser accelerator. It is a natural conclusion from the reported damage data that while the initial DLA work can be carried with more readily manufacturable silicon structures, a full realization of the DLA’s ultra-high gradient potential requires the development of nanofabrication methods for high band gap photonic materials, such as sapphire, which exhibit an order of magnitude higher breakdown thresholds. These results can be applied to other emerging applications of intense, picosecond, mid-IR lasers, such as HHG.

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