Gallium Oxide for High-Power Optical Applications

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Gallium oxide ($\text{Ga}_2\text{O}_3$) is an emerging wide-bandgap transparent conductive oxide (TCO) with potential applications for high-power optical systems. Herein, $\text{Ga}_2\text{O}_3$ fabricated nanostructures are described, which demonstrate high-power laser induced damage threshold (LIDT). Furthermore, the demonstration of an electron accelerator based on $\text{Ga}_2\text{O}_3$ gratings is reported. These unique $\text{Ga}_2\text{O}_3$ nanostructures provide acceleration gradients exceeding those possible with conventional RF accelerators due to the high breakdown threshold of $\text{Ga}_2\text{O}_3$. In addition, the laser damage threshold and acceleration performance of a $\text{Ga}_2\text{O}_3$-based dielectric laser accelerator (DLA) are compared with those of a DLA based on sapphire, a material known for its high breakdown strength. Finally, the potential of $\text{Ga}_2\text{O}_3$ thin-film coatings as field reduction layers for Si nanostructures is shown; they potentially improve the effective LIDT and performance of Si-based DLAs and other high-power optical structures. These results could provide a foundation for new high-power optical applications with $\text{Ga}_2\text{O}_3$.

Metamaterials are subwavelength structures that have recently enabled many new optical applications with a more compact form factor than traditional counterparts.[1–6] With the reduced feature sizes and scaling of these novel optical devices, many applications will achieve higher performance if nanostructures of $\text{Ga}_2\text{O}_3$ to demonstrate the first $\text{Ga}_2\text{O}_3$ nanostructures produced compact laser-driven electron accelerator. Being a TCO with high optical power tolerance could potentially make $\text{Ga}_2\text{O}_3$ an ideal candidate for both laser accelerator nanostructures (DLA) and other metamaterial applications, such as low-loss plasmonics.[3] In addition, we analyze the potential of $\text{Ga}_2\text{O}_3$ as a thin-layer coating for Si nanostructures to potentially enhance the performance of Si-based dielectric laser accelerators (DLA).

Figure 1a illustrates our laser-driven grating structure in which evanescent fields synchronously accelerate electrons by transferring energy from light to electrons. This structure utilizes a laser polarized parallel to the electron propagation direction normally incident upon the grating. For an electron accelerator, the acceleration gradient $G_{\text{acc}}$ is defined as the energy gain of the particles per unit distance is used to characterize the performance of the gratings.

$$G_{\text{acc}} = \frac{1}{\Lambda} \int_0^\Lambda E_z(\tau, t) d\tau = \frac{f_A}{\eta} E_{\text{inc}}$$

where $E_z(\tau, t)$ is the real-time in situ electric field experienced by the electrons in the direction of electron propagation. $f_A = G_{\text{acc}} / E_{\text{inc}}$ is called the field ratio, which is a geometry-dependent parameter independent of $E_{\text{inc}}$, where $E_{\text{inc}}$ is the amplitude of the incident laser electric field. For each given $E_{\text{inc}}$, there are one or more spots within the grating structure where the highest peak electric field amplitude, $E_{\text{max}}$, is located. The geometry-dependent enhancement factor is defined as $\eta = E_{\text{max}} / E_{\text{inc}}$. $E_{\text{threshold}}(\text{max}[E_{\text{max}}] = \eta \text{ max}[E_{\text{inc}}])$ is the maximum with a high damage threshold to handle high optical powers can be created. Such applications include: LIDAR components that enable higher laser power for longer range detection, flat lenses in laser-based defect inspection systems for the semiconductor industry, and dielectric laser accelerators (DLA) that have the potential to dramatically shrink both the size and cost of systems (including medical imaging and therapy, XUV lithography, etc.) centered around high energy particle beams.[7–13] Gallium oxide ($\text{Ga}_2\text{O}_3$) is a relatively newly developed wide bandgap semiconducting oxide with many distinct properties. $\text{Ga}_2\text{O}_3$ has demonstrated its potential as a transparent conductive oxide (TCO) for deep UV photodetectors and high-power electronics.[14–24] In this paper, we leverage the high laser damage threshold[25,26] and moderate conductivity of $\text{Ga}_2\text{O}_3$ to demonstrate the first $\text{Ga}_2\text{O}_3$ nanostructures produced compact laser-driven electron accelerator. Being a TCO with high optical power tolerance could potentially make $\text{Ga}_2\text{O}_3$ an ideal candidate for both laser accelerator nanostructures (DLA) and other metamaterial applications, such as low-loss plasmonics.[3] In addition, we analyze the potential of $\text{Ga}_2\text{O}_3$ as a thin-layer coating for Si nanostructures to potentially enhance the performance of Si-based dielectric laser accelerators (DLA).

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electric field amplitude within the structure just prior to optical field breakdown. Another important parameter, the damage factor \( DF = \left( 1 - \frac{f_A - \eta}{\eta - f_{\text{acc}}} \right) \), is used to characterize how effectively the gratings serve as a phase mask to utilize the incident electric field to accelerate electrons while not possess too many hot spots in the electric field distribution to survive a high electric field amplitude. For efficient coupling of the field to the electrons, the grating period is subject to the phase-matching constraint, \( \Lambda = \beta \lambda \), where \( \lambda \) is the drive laser wavelength and \( \beta \) is the electron velocity normalized to the speed of light, \( c \).

We use a finite-difference time-domain (FDTD) method to optimize both the field ratio, \( f_A \) and damage factor, DF. We optimize the width, \( w \) and depth, \( d \) for maximal acceleration gradient of Ga\textsubscript{2}O\textsubscript{3} DLAs at \( \beta = 0.54 \), assuming a laser wavelength of 2.0 \( \mu \)m. The optimized feature sizes have \( d = 600 \) nm and \( w/\Lambda = 50\% \). Both \( f_A \) and DF decrease by less than 10\% with tolerance windows of 600 \( \pm \) 100 nm and 50 \( \pm \) 10\% for depth and duty cycle, respectively.

Figure 1b shows the drive laser normally incident upon the fabricated grating device. The drive laser used in this experiment (indicated in red with the arrow showing the laser \( \vec{k} \) vector) has a wavelength of 2.0 \( \mu \)m, with a pulse duration of 250 fs and 100 kHz repetition rate. The electron beam travels vertically from top to bottom. Figure 1c shows a schematic of the laser-driven electron accelerator experimental setup. The pulsed electron beam is generated by UV laser pulses synchronized with the drive laser, which is incident on a flat copper photocathode, producing a \( 1 \pm 0.2 \) ps electron bunch. The electrons are focused by a magnetic lens to a spot size of 700 \( \pm \) 100 nm at the interaction point (the tip of the red arrow in Figure 1b). The electrons interact with the drive laser near the surface of the grating. The electron beam travels through the structure to a magnetic spectrometer and is incident on a microchannel plate (MCP) detector where the electron energy spectrum is measured with an energy resolution of 40 eV. The sapphire and Ga\textsubscript{2}O\textsubscript{3} nanostructures fabrication details are described in the Experimental Section. Figure 1d–f displays the SEM images of the sapphire and Ga\textsubscript{2}O\textsubscript{3} nanostructures. Zoomed-in views of each are shown in Figure 1e–g, respectively.

The electron spectra measured in the MCP for both sapphire and Ga\textsubscript{2}O\textsubscript{3} DLAs are shown in Figure 2a–d. The horizontal axis is longitudinal energy modulation with respect to the initial electron energy of 96.3 keV (\( \beta = 0.54 \)). The vertical axis is electron deflection in mrad. The “Laser Off” images in Figure 2a,b show the distribution of electron density in the absence of the
laser–electron interaction. In contrast, the “Laser On” images (Figure 2c,d) show two distinct tails on each side of the distribution. These tails are composed of decelerated and accelerated electrons. From the charge density shown in Figure 2a–d, we plot the normalized electron counts as a function of energy modulation in Figure 2e,f. The depletion of the electron density in the central energy peak for the laser on curve (in red), when compared to the laser off curve (in blue) indicates that electrons initially at this central energy experienced energy gain or loss due to interaction with the drive laser. The maximum energy gain of the sapphire DLA was 1\(\pm\)0.1 keV over an interaction distance of 18\(\mu\)m, which gives an acceleration gradient of 56\(\pm\)6 MeV m\(^{-1}\) at an \(E_{\text{inc}}\) of 3.1 GV m\(^{-1}\). The maximum energy gain of the Ga\(_2\)O\(_3\) DLA is 2.4\(\pm\)0.1 keV over an interaction distance of 23\(\mu\)m, producing an acceleration gradient of 104\(\pm\)4 MeV m\(^{-1}\) at an \(E_{\text{inc}}\) of 2.4 GV m\(^{-1}\).

These are the first successful demonstrations of laser-driven electron acceleration with wide-bandgap semiconducting Ga\(_2\)O\(_3\) nanostructures. The acceleration gradient of this grating-based Ga\(_2\)O\(_3\) DLA is nearly double that of the sapphire structure. We hypothesize that the higher conductivity of Ga\(_2\)O\(_3\) allows a closer approach of the electrons to the grating surface, resulting in higher acceleration fields due to the exponential decay nature of the nearfield in the gratings.\(^{8,9,27}\) The measured LIDT is 0.36\(\pm\)0.04 J cm\(^{-2}\) for Ga\(_2\)O\(_3\) gratings both in vacuum and in air, which is comparable to that of sapphire gratings (0.40\(\pm\)0.04 J cm\(^{-2}\)). The maximum \(E_{\text{inc}}\) before damage occurs is thus 3.3 GV m\(^{-1}\) for Ga\(_2\)O\(_3\) gratings, using laser parameters of a wavelength of 2.0 \(\mu\)m, with a pulse duration of 250 fs and 100 kHz repetition rate. (LIDT measurements are described in the Experimental Section). The Ga\(_2\)O\(_3\) DLA demonstrated here was

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Figure 2. a) Laser off charge spatial distribution of sapphire DLA. b) Laser off charge spatial distribution of Ga\(_2\)O\(_3\) DLA. c) Laser on charge spatial distribution of sapphire DLA. d) Laser on charge spatial distribution of Ga\(_2\)O\(_3\) DLA. e) Normalized electron counts as a function of electron energy modulation of sapphire DLA. f) Normalized electron counts as a function of electron energy modulation of Ga\(_2\)O\(_3\) DLA.
Silicon is, by far, the most developed, least expensive and largest wafer diameter material that enables fabrication of complex, precise nanostructures. Fortuitously, it is also a very efficient phase mask at wavelengths from the visible to mid-infrared, due to its comparably high index of refraction. Therefore, we propose to leverage the higher LIDT of Ga$_2$O$_3$ discussed above to improve Si nanostructures performance. This could be achieved by adding a thin-film Ga$_2$O$_3$ coating to a Si grating structure, which acts as a field reduction layer (Figure 3a,b). The reduction of the local field enhancement at the sharp edges is notable. Additionally, the ideal material for a Si AR coating has an index of $\approx 1.85$. Ga$_2$O$_3$ has a refractive index of 1.9 in NIR. Here, we choose a 30 nm Ga$_2$O$_3$ conformal coating layer for the Si grating. 30 nm is chosen as a trade-off between the following factors: 1) Thicker films (such as quarter-wave layers) provide better antireflection (AR) coating, but will markedly reduce the $f_A$ and thus lead to lower $G_{acc}$. 2) Thinner films do not provide a significant field reduction.

Figure 3a shows the calculated electric field distribution near a Si grating structure designed for a 2.0 $\mu$m drive laser wavelength. One can see from Figure 3b that by adding a 30 nm Ga$_2$O$_3$ coating, the field enhancement factor $\eta = E_{\text{max}}/E_{\text{inc}}$ is reduced by 2.9 times (comparing coated vs noncoated samples). The hotspots now occur at the surface of the Ga$_2$O$_3$ instead of the Si surface, and thin-film Ga$_2$O$_3$ could potentially tolerate a higher $E_{\text{inc}}$ than the bulk Si structure. In theory, if the LIDT is determined only by the incident electric field amplitude, given $\eta = E_{\text{max}}/E_{\text{inc}}$ is reduced by 2.9 times with an $f_A$ reduction of less than 1% by adding this thin layer of Ga$_2$O$_3$ illustrated in the FDTD simulation, $DF = f_A/\eta = G_{acc}/E_{\text{max}}$ increases by 2.9 times as shown in Figure 3c,d.

In one set of our LIDT measurements, we found that no change of LIDT (0.04 J cm$^{-2}$) was observed when using a 250 fs, 100 kHz repetition rate, 2 $\mu$m wavelength laser. In another set of experiments with 100 fs, 1 kHz repetition rate, 2 $\mu$m wavelength laser, max{$E_{\text{inc}}$} increases from 0.83 GV m$^{-1}$ (Si DLA) to 1.52 GV m$^{-1}$ (coated Si DLA). In the first set of experiments with a higher repetition rate and longer pulse duration of 250 fs, we believe that thermal effects, such as thermal melting, contribute to the laser-induced damage. In the second set of experiments, we believe that the damage is likely field induced damage with reduced thermal effects because of the lower, 1 kHz repetition rate. These results merit further laser damage studies and better film quality development in the future.

![Figure 3](image-url)  
**Figure 3.** a) Peak electric field distribution of Si DLA. b) Peak electric field distribution of Si DLA with a 30 nm conformal Ga$_2$O$_3$ thin film coating layer. The effect of this treatment is twofold: the edges are rounded, which reduces the peak field, while the LIDT is also larger in the coating layer. c) DF of the Si DLA as a function of grating depth and duty cycle. d) DF of the Si DLA with a 30 nm Ga$_2$O$_3$ coating as a function of grating depth and duty cycle.
In summary, we demonstrated Ga2O3 fabricated nanostructures with a high laser induced damage threshold (LIDT) of 0.36 ± 0.04 J cm−2, comparable to that of sapphire. Furthermore, we report the demonstration of an electron accelerator based on Ga2O3 nanostructures, which provides an acceleration gradient of 104 ± 4 MeV m−1, exceeding those possible with conventional RF accelerators due to the high breakdown threshold of Ga2O3. In addition, we show the potential of thin-layer coatings as field reduction layers for Si nanostructures to potentially improve the effective LIDT and performance of Si-based nanostructures. These results demonstrate the promising high-power optical application of Ga2O3 nanostructures as a miniaturized laser-driven electron accelerator. With the distinct electrical and optical properties of Ga2O3 combined with advances in fabrication and wafer growth techniques, more novel high-power optical applications based on Ga2O3 will be realized in the near future.

Experimental Section

DLA Fabrication: Sapphire gratings were fabricated on 4 in. c-plane sapphire substrates at the Stanford Nanofabrication Facility (SNF). Nanopatterning was performed with conventional optical lithography using an ASML PAS5500 with Shipley 955 photoresist and inductively coupled plasma (ICP) etching with BCl3 using Oxford Instruments PlasmaPro System 100 Cobra III-V Etcher (Ox-35). This ICP-etching recipe used 15 sccm of BCl3 at a chamber pressure of 3 mTorr with 600 W ICP power and 150 W of bias, which led to an etch rate of 50 nm min−1 with a selectivity between sapphire and Shipley 955 photoresist to be 1:1.7. The grating was fabricated on top of a 6 µm high mesa structure that minimizes electron beam clipping on the substrate edges. The grating period was $\Lambda = 1080$ nm, with a grating depth $d = 600$ nm and duty cycle $w/\Lambda = 50\%$. $\Lambda = 1080$ nm, which was synchronous with 96.3 keV ($\beta = 0.54$) electrons when driven with a 2.0 µm laser.

The Ga2O3 gratings were fabricated on 2 in. $\beta$-Ga2O3 substrates at the SNF and Stanford Nano-Patterning Center (SNC). The β-Ga2O3 (~201) wafers (purchased from Tamura Corporation in Japan) obtained by edge-defined film-fed growth (EFG) were unintentionally doped (UID) with 0.09 Ω cm measured by Tamura Nanofabrication. Nanopatterning was performed by electron beam lithography using a JEOL JBX-6300 with ZEP 520a e-beam resist and ICP etching with BCl3 using Ox-35. This ICP-etching recipe used 15 sccm of BCl3 at a chamber pressure of 3 mTorr with 600 W ICP power and 150 W of bias, which led to an etch rate of 50 nm min−1 with a selectivity between β-Ga2O3 and Shipley 955 photoresist to be 1:1.67. The grating was fabricated on a 10 µm high mesa structure. The grating period $\Lambda = 1080$ nm, with the grating depth $d = 580$ nm and duty cycle $w/\Lambda = 50\%$.

The Ga2O3 films were deposited on Si by Veeco Savannah thermal ALD system in SNF using a precursor of Ga(NEt3)3 and DI water steam at 200 °C with a deposition rate of 0.6 Å s−1. The pulse durations for Ga(NEt3)3 and DI water steam were 0.4 s and 0.015 s, respectively. The purge time was 30 s between two pulses. The uniformity of this Ga2O3 ALD film was verified by Woollam M2000 ellipsometer to be 30 ± 0.5 nm across a 2 in. wafer.

DLA LIDT Measurements: LIDT measurements were conducted to determine the maximum “safe” fluence, i.e., the reported LIDT was the minimum fluence value at which we had observed damage. Although devices can be selected to mitigate the stochastic distribution of defects in Ga2O3, it is most important to find this minimum damage fluence in a DLA experiment and many similar optical applications so that this highest “safe” fluence can be chosen for operation. LIDT studies were performed on 12 identical grating devices, at multiple locations per device. For each device, the minimum damage fluence value was recorded as the LIDT. The devices were illuminated for 100 s by a 100 kHz (or 1 kHz) laser focused at the surface of the grating with a 1/e2 radius of 20 ± 2 µm. After each 100 second exposure, an imaging microscope was used with 1 µm resolution to visually inspect for damage. Any visible change in optical properties of the grating observed by optical inspection was considered “damage.” If the device was undamaged, the fluence was increased by roughly 2% of overall LIDT and the measurement repeats until damage occurred. The uncertainty of the reported LIDT was derived from the laser pulse energy uncertainty.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords
dielectric laser accelerator, gallium oxide, high-power optical systems, lasers, nanostructures, optical materials

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