

# Optics Letters

## Harmonic generation in solids with direct fiber laser pumping

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Received 19 January 2017; accepted 13 February 2017; posted 17 February 2017 (Doc. ID 285020); published 9 March 2017

**High harmonic generation in solids presents the possibility for bringing attosecond techniques to semiconductors and a simple source for frequency comb spectroscopy in the vacuum ultraviolet. We generate up to the seventh harmonic of a Tm fiber laser by focusing in silicon or zinc oxide. The harmonics are strong and stable, with no indication of material damage. Calculations show the potential for generating nineteenth harmonic photons at 12 eV photons of energy.** © 2017 Optical Society of America

**OCIS codes:** (190.2620) Harmonic generation and mixing; (190.4720) Optical nonlinearities of condensed matter; (190.7110) Ultrafast nonlinear optics.

<https://doi.org/10.1364/OL.42.001113>

High harmonic generation in gases is an important technique for generating attosecond pulses and studying molecular structure and dynamics. It arises when an electron tunnel ionizes, accelerates in the laser field, and then recollides with the ion, releasing its kinetic energy as a high harmonic photon [1]. The high ionization potential of gas molecules means that intensities over  $10^{14}$  W/cm<sup>2</sup> are needed to produce useful amounts of high harmonic radiation, which is generally achieved using room-sized, multi-stage Ti:sapphire amplifier systems [2].

There are two methods for reducing the laser demands of high harmonic generation. One is to use optical enhancement cavities to recycle pulses such as from high-repetition-rate fiber lasers, which have high average power, but low pulse energy, increasing the intensity inside the cavity enough to generate high harmonics [3,4].

Another method is to replace the gas with a solid [5], where the ionization potential becomes a bandgap that can be changed by choice of material. Photons at 62 nm, or 20 eV, have been generated in the large bandgap material MgO using a 1.3  $\mu$ m wavelength driver [6]. For ZnO, a semiconductor with a 3.3 eV bandgap, intensities of  $10^{12}$  W/cm<sup>2</sup> have generated almost 10 eV photons at the 25th harmonic [5]. The smaller bandgaps of ZnO and important semiconductors such as silicon add another demand on the laser, that the wavelength be well into the near infrared to avoid multiphoton absorption.

Previous experiments with high harmonic generation in solids have used Ti:sapphire systems with additional nonlinear stages such as optical parametric amplification (OPA) and difference frequency generation to reach longer wavelengths [7,8], further complicating the laser source.

Here, we take the natural next step in this progression and combine a fiber laser with a solid, removing the complications of using an enhancement cavity. Our thulium laser directly provides high-power femtosecond pulses at 1.9  $\mu$ m for harmonic generation in silicon or zinc oxide. With a simple fiber laser, we generate up to the seventh harmonic, about 270 nm wavelength, with an order of magnitude less cost than a mJ Ti:sapphire-pumped OPA.

Thulium fiber lasers can be easily made into frequency combs [9], making this a potential source of practical ultraviolet frequency combs for new applications such as high-resolution electronic transition spectroscopy and next-generation clocks using nuclear resonances [4]. Driving harmonic generation with simpler lasers is also a necessary step toward realizing practical applications of attosecond physics and electronics [10].

In these experiments, we focus the pulses from a thulium fiber laser into Si or ZnO samples and measure the resulting harmonics with a spectrometer or a CCD array. The thulium laser provides 60 fs pulses at a 93.4 MHz repetition rate with up to 1 W average power. The pulses have good mode quality, since they emerge directly from standard polarization-maintaining 1550 nm optical fiber, particularly compared to solid-state amplifiers.

The Tm amplifier is seeded by pulses from a polarization-maintaining erbium oscillator, similar to [11], which is broadened out to the 2  $\mu$ m wavelength region by supercontinuum generation. Seeding the amplifier with a thulium oscillator would provide similar performance [9].

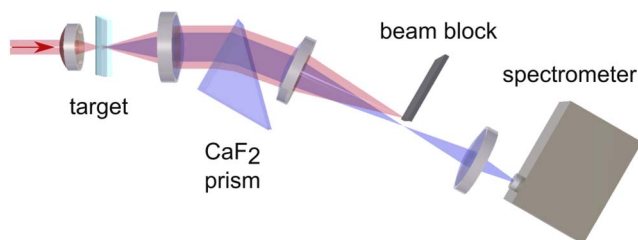
To keep the focusing optics simple, we used identical 4 mm effective focal length chalcogenide aspheric lenses for both collimation from the fiber and focusing onto the sample. This symmetric arrangement is intended to reduce aberrations and provide a focus comparable to the fiber mode. We measured the mode size by a knife edge, finding a 90/10 diameter of about 8  $\mu$ m, but this value is quite sensitive to alignment. The peak intensity in these conditions would be

over  $10^{11}$  W/cm<sup>2</sup>, but chromatic aberrations likely increase the pulse duration. Since there is further optimization of optical alignment while observing third harmonic generation, the final beam size may be smaller.

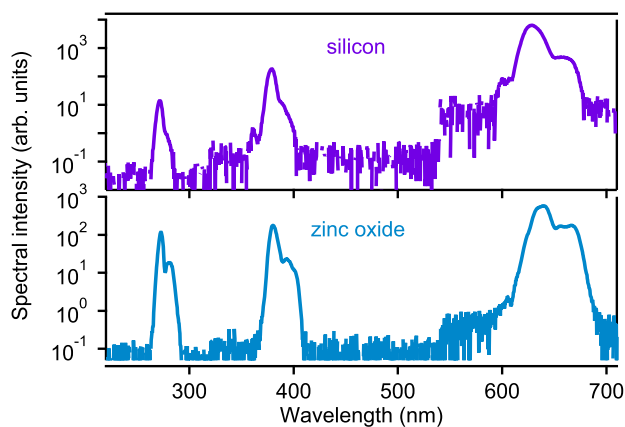
Figure 1 illustrates the collection optics. After the sample, light is collimated by a 20 mm focal length CaF<sub>2</sub> lens. Since this still includes the strong pump beam, we spatially separate the wavelengths with a CaF<sub>2</sub> prism and lens, blocking the pump beam at the focus. We then refocus into the  $\sim 100$   $\mu$ m slit of a CCD grating spectrometer. Different wavelengths focus at different positions at the spectrometer slit as well, resulting in different optimal spectrometer positions for each harmonic. This has the benefit of letting us measure a weak harmonic, while reducing entry of the much brighter third harmonic into the spectrometer, which would otherwise overwhelm the weaker signals. However, this complicates comparisons between harmonics, requiring more than one measurement to acquire a full bandwidth spectrum.

To generate harmonics, we used two samples: a 120 nm monocrystalline silicon film on 0.5 mm of sapphire and a 200  $\mu$ m thick monocrystalline zinc oxide disk. We confirmed that a 0.5 mm thick plain sapphire window only produced detectable third harmonic.

Concatenated harmonic spectra from the two samples are shown in Fig. 2. The third harmonic is strong and easily visible



**Fig. 1.** Layout of experiment optics. Harmonics are generated by focusing into a silicon or zinc oxide target and separated by a prism to block the strong pump beam or strong low-order harmonics from reaching the spectrometer.



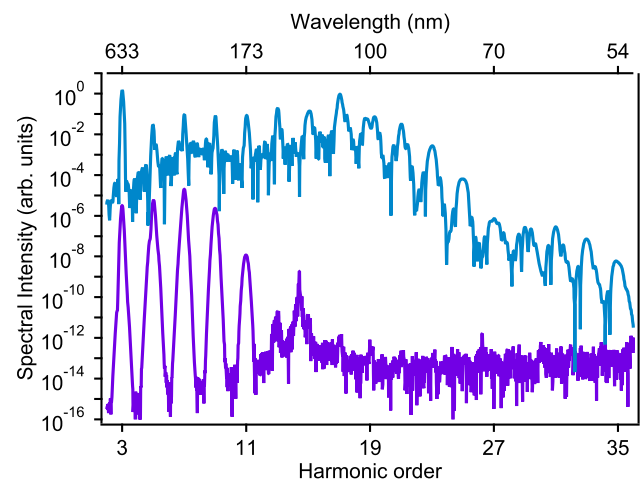
**Fig. 2.** Spectra of the seventh, fifth, and third harmonics of a 1 W thulium fiber laser generated in thin-film silicon and zinc oxide. Silicon has a 1.1 eV indirect, and 3.5 eV direct bandgap. Zinc oxide has a 3.3 eV direct bandgap.

by eye. The fifth and seventh harmonics were strong, with the seventh harmonic from zinc oxide saturating the spectrometer in a few milliseconds. We did not detect any ninth harmonic of the thulium pump, the highest harmonic within our spectrometer range.

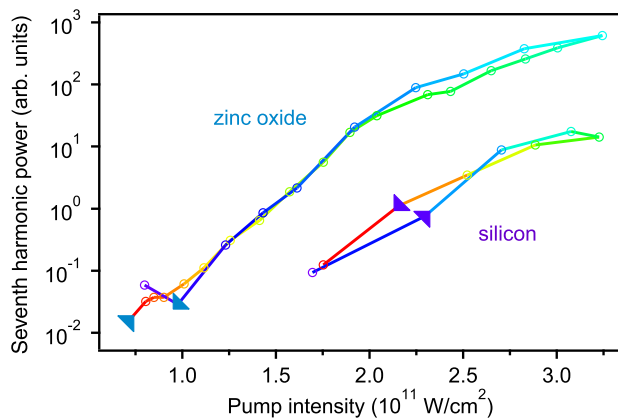
We calculated the expected harmonic generation for the case of a  $6.6 \times 10^{11}$  W/cm<sup>2</sup> peak intensity before entering the material. This intensity is likely higher than our actual intensity due to aberrations, and may be taken as an upper bound for predicted harmonic generation. Semiclassical modeling of an electron-hole pair [12,13] resulted in a harmonic cutoff at harmonic 7.8. A quantum calculation of the expected harmonic spectrum resulted in a cutoff at harmonic 9, as shown in the purple, lower curve in Fig. 3. The quantum calculation predicts the generation of higher orders because it includes the offset of the electron birth position relative to the hole, which is not part of the semiclassical calculation. The blue, upper curve shows a calculation for stronger pumping of ZnO, with 6.25 times more incident intensity and an anti-reflection coating, indicating the potential for generating nineteenth harmonic photons at 100 nm wavelength, or 12 eV of photon energy. Such higher intensities should be possible by moving to large-core fiber to reduce nonlinearities from higher pulse energies.

Material damage from the high laser fluence was expected to be a limiting factor when using a high-repetition-rate fiber laser. In our experiments, we see obvious thermal effects, but no noticeable permanent damage. From our experience with the higher pulse energy Ti:sapphire system, we suspect that zinc oxide may tolerate a six times higher intensity, but silicon would likely be damaged.

One way we test for damage from the thulium laser is to measure the harmonic output while stepping up the pumping power with a neutral density gradient filter, illuminating for tens of seconds at each step, then stepping back down. Figure 4 shows the spectrometer signal integrated over the seventh harmonic as a function of the estimated pump intensity. Since the harmonic production is not found to be lower when the intensity is stepped down, we take this to mean that the material is not being damaged by even the full pump intensity.



**Fig. 3.** The purple, lower curve is a quantum calculation of the harmonic spectra from silicon near our experimental conditions. The blue, upper curve is for ZnO with a 6.25 times higher pump intensity and antireflection coating, indicating the possibility for photon generation in the 10 eV range.

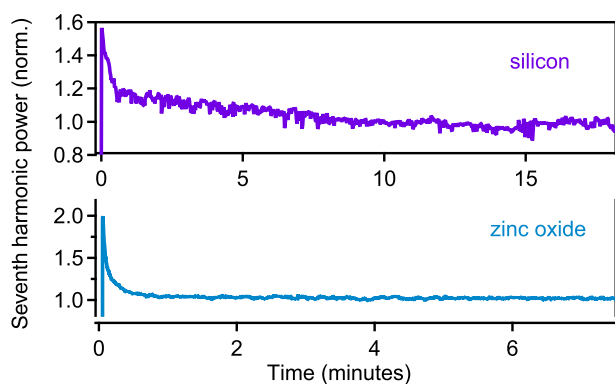


**Fig. 4.** Seventh harmonic signal from zinc oxide and silicon integrated over the spectrum as the pump power is stepped up and back down, with direction indicated by the arrows. The repeatability in both directions indicates that the sample is not being damaged by the strong pump.

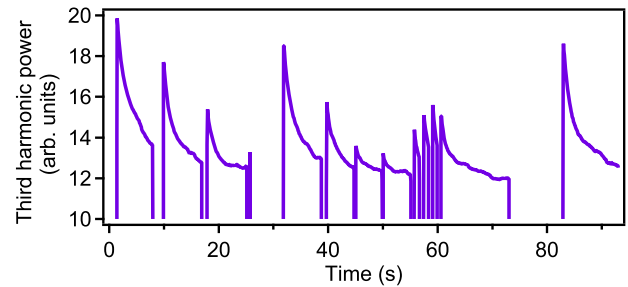
A second test we performed was to record the harmonic output over a length of time, as in Fig. 5. The signal changes with a timescale of seconds when first illuminated, then reaches a steady state, which we interpret as thermal heating and stabilization. The initial decay may appear to be from damage, but there are cases where the harmonic output increases upon illumination, depending on optical alignment.

For a third test, we modified the previous test by replacing the spectrometer with a large CCD array, and by blocking and unblocking the pump beam several times. Using a CCD removes thermal lensing effects from the collection process by acting as a large area detector. The harmonic output is taken to be the integral of a large image region around the laser spot, avoiding alignment issues from focusing into a spectrometer slit. We reduce pump light to the camera by reflection from an 850 nm longpass filter. Light pickup from the room is reduced by transmission through a red filter and a neutral density filter, leaving mostly third harmonic to reach the camera.

Figure 6 shows an example of this integrated CCD signal with time. The signal is consistent with heating, which can



**Fig. 5.** Seventh harmonic signal from zinc oxide and silicon integrated over the spectrum, taken over several minutes. There are early changes at thermal timescales, but the signal remains consistent, indicating either no damage, or no continuing damage.



**Fig. 6.** Third harmonic signal from silicon with time, while the pump beam is blocked for different periods. The decay and recovery of the output depends on the blocking time, consistent with heating from the pump, rather than accumulating damage.

cause increased light absorption by thermal carrier generation. When the beam is blocked for short times, there is still a fast change in the signal, but the overall signal quickly returns to its previous path, while longer blocking resets the behavior. This is consistent with fast heating and cooling at the focal spot within seconds, and slower heating and cooling in the surrounding crystal on the order of 10 s. The output recovery after blocking confirms that the fast decay is not permanent damage. Visual inspection of the samples with a conventional microscope showed no damage.

Harmonic generation in solids is at the intersection of strong-field, attosecond, frequency comb, and semiconductor science. Strong fields in semiconductors require long wavelengths to stay below the bandgap, making them a good match for thulium frequency comb fiber lasers.

As a medium, solids can be much thinner than a gas jet, reducing issues such as the Guoy phase shift in tight focuses. Semiconductor techniques are highly advanced, allowing the engineering of many details, from doping to nanostructures [14], which can serve for enhancing the output as frequency comb sources or developing new methods in attosecond electronics. For example, we have seen with our Ti:sapphire-pumped OPA that gold nanoantennas on silicon, resonant at 2  $\mu\text{m}$  wavelength, can enhance the high harmonic emission by 10 times [15,16], a technique that should be particularly useful for improving the effective intensity of fiber laser pumping.

We have shown here a key step in expanding research and application of harmonic generation in solids by greatly simplifying the laser system requirements. Our femtosecond thulium laser generates up to the seventh harmonic in silicon and zinc oxide without causing material damage. Neither the laser nor the target was optimized for this experiment. Simple changes such as designing the laser for high pulse energy, or cooling and anti-reflection coating the target will yield large improvements in the output. The generation of harmonics in solids by fiber lasers is a promising technique, providing short wavelength, coherent pulses at MHz repetition rates for new applications such as ultrafast imaging and diffraction.

**Funding.** Air Force Office of Scientific Research (AFOSR) (FA9550-15-1-0037, FA9550-16-1-0109); National Research Council Canada (NRC); Canada Research Chairs; Natural Sciences and Engineering Research Council of Canada (NSERC); Canada Foundation for Innovation (CFI); Ontario Research Fund.

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