

Coherent Extreme Ultraviolet Source using a Single Dielectric Nanostructure

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High-harmonic generation (HHG) using crystalline solids is a novel method to generate coherent extreme-ultraviolet pulse. Metal based nanostructure using plasmonic field enhancement can improve the efficiency of HHG. However, the metal based nanostructures used for plasmonic HHG have a limitation on low damage threshold of metal. This study demonstrates an all-dielectric conical sapphire nanostructure used as a compact HHG emitter that generates high order harmonics with wavelengths up to approximately 60 nm without severe damage. We compare the structure with a gold layered conical sapphire nanostructure. The conical sapphire structure has a higher damage threshold and reusability for EUV generation even though it has a lower HHG intensity than the gold layered conical sapphire structure because of the lower intensity enhancement. The results confirm the possibility of a compact EUV light source for nanoscale applications.

1. Introduction

High-harmonic generation (HHG) is a key technique for generating coherent ultrashort pulses in extreme-ultraviolet (EUV) and X-ray bands by an interaction between the high-power laser pulse and the material's electrons [1,2]. The ultrashort pulses based pump-probe technique can measure ultrafast dynamics with high temporal resolution.

HHG was explained by the tree-step model and it has been reported in atomic gas [3], crystalline solids [4,5], liquids [6], and plasma [7]. When the electric field amplitude of the laser pulse is comparable to the electric field in atoms, electrons can be ionized by tunneling effect. The detached electron is accelerated by the laser field and is recombined with its parent ion. To replace a large and complex high power laser system for inducing HHG, plasmonic nanostructure was selected for amplifying a driving femtosecond laser field and initiating HHG [8]. The nanostructure converted NIR femtosecond laser pulse to EUV pulse without strong fluorescence signals. However, the enhanced field and thermal energy deformed the metal-dielectric nanostructure and the deformation diminished HHG efficiency.

In this study, we designed and fabricated a single conical dielectric nanostructure using theoretical calculation and semiconductor fabrication facilities. The structure compared with the metal-sapphire based nanostructure based on the HHG in the EUV band and the damage results according to the field enhancements.

2. Theoretical Calculation and Experiments

2.1 Simulation

A single conical sapphire nanostructure enhanced driving laser field by field enhancement and internal reflection. The enhanced laser field induce nonlinear dynamics of electrons at the end of the nanostructure. HHG is result of the nonlinear electron dynamics. Figure 1b indicates a single conical sapphire nanostructure prepared to HHG for EUV generation. The scanning microscopy (SEM) image of the conical sapphire nanostructure shows its shape and dimensions. The nanostructure has a height of 1.5 μm , and a base diameter of 2.6 μm . The nanostructure was fabricated by plasma dry etching on a flat single-crystalline sapphire wafer (C-plane) with a thickness of 430 μm .

Field enhancement of the conical sapphire nanostructure was calculated based on the finite-difference time-domain (FDTD) calculation. The structure was modeled with cubic elements of $5.0 \times 5.0 \times 5.0 \text{ nm}$. A boundary condition of the calculated area was set as a perfectly matched layer to minimize the reflections and other effects. A truncated cone was selected for the conical sapphire nanostructure. The driving laser pulse was designed having a 12-femtosecond pulse duration and an 800 nm center wavelength. The driving laser pulse was incident on the bottom of the nanostructure and propagate inside the nanostructure. The polarization of the driving laser pulse is parallel to the X-axis of the cross section (Fig. 1c). The intensity enhancement was defined as $|E/E_0|^2$. E denotes the amplitude of the enhanced electric field, and E_0 is the amplitude of the driving femtosecond laser field. The maximum intensity enhancement occurred inside the center

of the conical structure and reached approximately 50. Along the surface of the conical structure, substantial intensity enhancements occurred periodically, reaching approximately 6.8 at the top surface.

Since there was no metal-dielectric interface, there was no plasmonic field enhancement as in previous research [9]. The enhancement comes from a combination of the electrostatic lightning rod effect due to the geometric distinctiveness of sharply pointed structures and an overlapped driving pulse that depends on the internal reflection inside the structure.

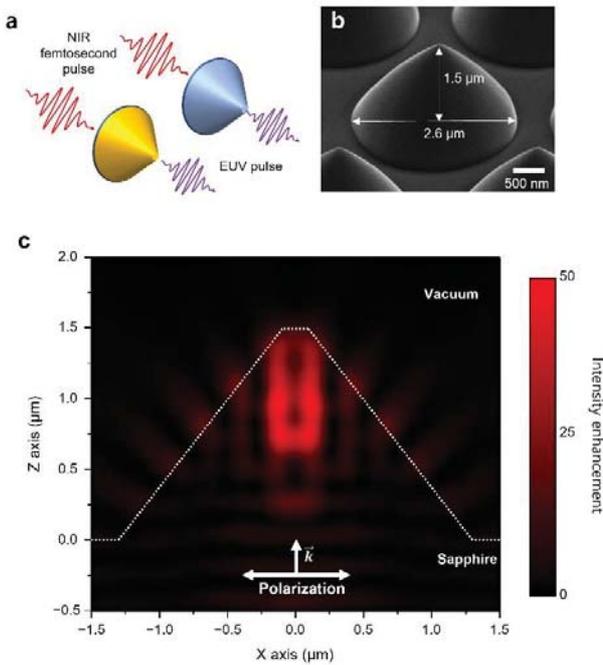


Fig. 1 (a) Concept of EUV nanogenerator using plasmonic field enhancement along with the conical sapphire nanostructure and the gold layered conical sapphire nanostructure. (b) SEM image of the conical sapphire nanostructure. (c) Theoretical calculation of the intensity enhancement of the conical sapphire nanostructure on the cross section.

2.2. Experimental Setup

12 fs pulses with 75 MHz repetition rate from a Ti:sapphire oscillator was focused to the bottom of the conical sapphire nanostructure. Chirp mirrors were used to maintain the short pulse duration of 12 fs on the sample. The half-wave plate and the polarizer control the input pulse energy keeping a polarization direction. An achromatic triplet lens focused the laser pulses with a spot size of 5 μm. The polarization direction of the incident laser field was aligned parallel to the sapphire C-plane and perpendicular to the sapphire A-plane. A toroidal mirror collected the generated EUV radiation emitted from the sample and delivered it to the entrance of the EUV spectrometer. A microchannel plate coupled with a phosphor screen and optical charge-coupled device measure EUV photon signal along a Rowland circle. The pressure of the entire vacuum chamber was maintained at approximately 10^{-6} mbar during the experiments.

3. Results and Discussion

Figure 2 shows the measured intensity of the high-order harmonic from the conical sapphire structure with increasing the driving laser intensity. The peak power intensity inside the material of the driving laser pulse was increased from 0.05 to 0.89 TW/cm². High harmonics up to the 11th order were observed when the focused intensity reached 0.89 TW/cm². HHG from bulk sapphire in a multi-photon regime follows perturbative scaling, given by $I_{HHG} \propto I^N$, where I_{HHG} denotes the intensity of the measured harmonic peaks, I is the peak intensity of the driving pulse, and N denotes the order of the harmonics [5]. The intensity of the harmonics from the conical sapphire structure behaved non-perturbatively with the laser intensity, similar to the results obtained from the ZnO nanocone [10].

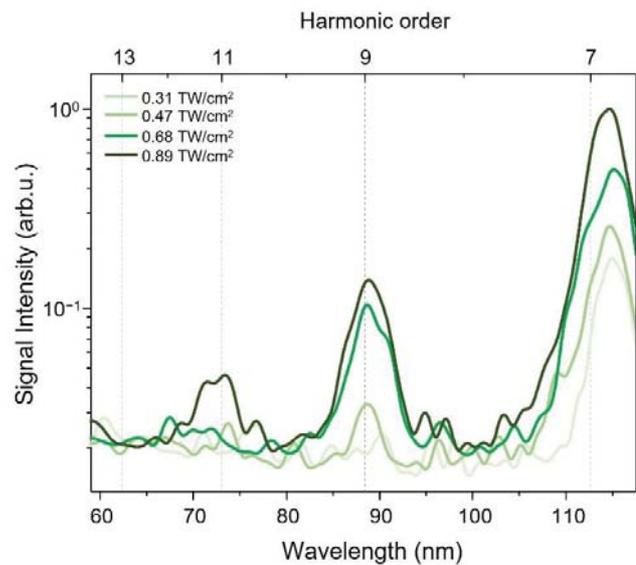


Fig. 2 HHG from a conical sapphire nanostructure. Measured EUV spectra with increasing intensity of the driving femtosecond laser.

The measured spectra from the gold layered conical sapphire structure and the conical sapphire structure are shown in Figure 3. The incident laser intensity was estimated to be 0.42 TW/cm² in the materials. Both nanostructures amplify the driving laser fields and generate high-order harmonics in the EUV band. The gold-layered sapphire funnel structure generates harmonics up to the 13th order, and the conical sapphire structure radiates up to the 9th order. The intensity of H7 from the gold layered conical sapphire structure is six times higher than that from the conical sapphire structure. The profile of the intensity enhancement from the tip of the conical sapphire structure is different from that of the gold layered conical sapphire nanostructure [8]. The average intensity enhancement of the tip of the conical sapphire structure was approximately 6.8, and the enhancement factor was uniform along the tip surface. The gold-layered conical sapphire nanostructure has an irregular profile with a higher intensity enhancement factor. The maximum enhancement factor was approximately 20 dB at the edge of the tip. The higher intensity enhancement of the gold-layered conical sapphire nanostructure led to a higher intensity and cutoff extension of the harmonic generation under identical conditions.

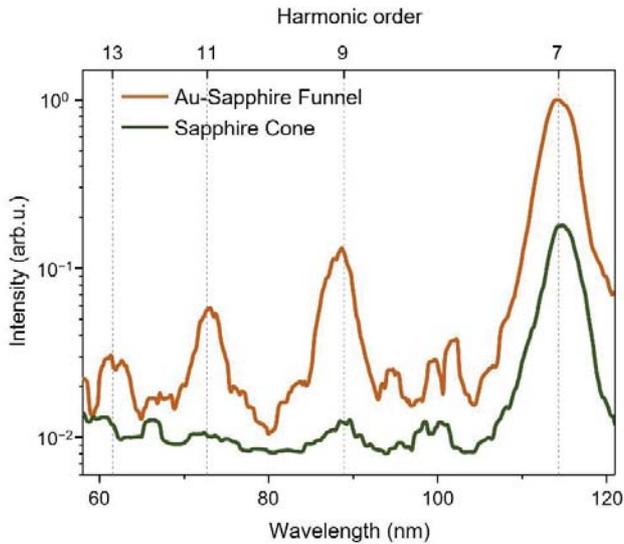


Fig. 3 Measured spectra in EUV band from the conical sapphire nanostructure and the gold layered conical sapphire nanostructure.

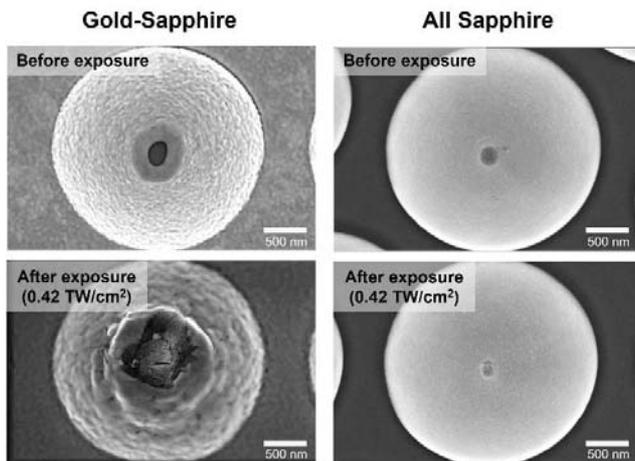


Fig. 4 SEM images for comparison between the conical sapphire nanostructure and the gold layered conical sapphire nanostructure. The gold layered conical sapphire nanostructure before laser exposure (left top) and after laser exposure (left bottom). The conical sapphire nanostructure before laser exposure (right top) and after laser exposure (right bottom). The laser exposure time is 30 s.

Figure 4 compares the gold layered conical sapphire nanostructure and the conical sapphire nanostructure. The SEM images were captured before and after a laser exposure of 0.42 TW/cm^2 . The SEM image of the conical sapphire structure shows no evident damage compared with that of the structure before laser exposure. Conversely, the gold layered conical sapphire nanostructure showed structural damage clearly. The gold layer was deformed by melting and vaporizing. The surface cracks were formed perpendicular to the direction of the incident laser polarization, which is shown along the Y -axis on the SEM image. A substantial field enhancement of the gold layered conical sapphire nanostructure causes thermal damage even for weak input laser intensities of $\sim 0.1 \text{ TW/cm}^2$. The deformed structure has reduced field enhancement, and the EUV yield from the structure varies with the exposure time. The conical sapphire structure had a structural change at the apex, but the

EUV harmonics were generated repeatedly under the experimental conditions.

4. Conclusion

This study demonstrated durable and efficient HHG in the EUV band using a single conical sapphire nanostructure. The experiments performed in this investigation demonstrated the conical sapphire structure as an effective emitter that produces coherent EUV harmonics at wavelengths up to $\sim 60 \text{ nm}$ for laser intensities of 1.13 TW/cm^2 with nJ-energy pulses emitted from a Ti:sapphire femtosecond oscillator. By comparing the conical sapphire structure with the gold layered conical sapphire nanostructure, we demonstrated the different EUV harmonic yields according to the field enhancements, and it was observed that the conical sapphire structure had a higher damage threshold and was reusable under the experimental conditions. The all-dielectric nanostructure with good durability and field enhancement can boost nonlinear processes on a table-top scale without an external laser amplification system.

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