Supplementary Materials

Generating ultrabroadband deep-UV radiation and sub-10 nm gap by hybrid-morphology gold antennas

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# Laser Physics and Nonlinear Optics, Mesa+ Institute for Nanotechnology, University of Twente, 7500AE Enschede, The Netherlands
Section I: Stability of plasmonic metasurface in strong femtosecond fields

We performed long-term measurements of third harmonic emission at two different driving laser intensities. At peak intensity of $\sim 50$ GW/cm$^2$, which was applied to perform the dispersion-scan measurement, the harmonic signal at 255 nm stays stable for half an hour, as shown in Fig. S1(a). This indicates that the metasurface can withstand such an intensity level for the duration of the dispersion-scan measurement and will also be practical for other long-term applications. When increasing the incident peak intensity to $\sim 600$ GW/cm$^2$, which was employed to modify the nanostructure for visualization of near-field enhancement map, the harmonic signal at 255 nm exponentially decays over time, as shown in Fig. S1(b).

![Figure S1](image)

**Figure S1**: Time evolution of THG harmonic signal at 255 nm when excited by a peak intensity of $\sim 50$ GW/cm$^2$ (a) and $\sim 600$ GW/cm$^2$ (b), respectively.

Section II: Estimation of third harmonic conversion efficiency

In the spectral region of interest, i.e., from 200 to 300 nm, the diffraction efficiency of our concave grating and quantum efficiency of our photomultiplier tube are both $\sim 20\%$. Considering these efficiencies, the calibrated spectra counts of the third harmonic radiation from the bare crystal and from our plasmonic metasurface are plotted in Fig. S2. The harmonic emission spectrum from the bare crystal is also calibrated by the gold-free area (i.e., 25% of
the surface of the sample is not covered by a gold film, characterized through the high-resolution SEM images). We observe that in addition to the harmonic enhancement by the plasmonic metasurface, the harmonic bandwidth is also evidently broadened. Our detection scheme has a down time which can decrease the detected photon numbers below the detector’s efficiency, by a non-quantifiable amount which gets worse closer to saturation (although we are far from saturation). Nonetheless, we can estimate a lower bound of the conversion efficiency. At an incident laser intensity of ~50 GW/cm², the total photon counts over the entirety of the third harmonic bandwidth from the nanoantennas is measured to be $1.7 \times 10^7$ photons/s. The photon energy at this spectral range is ~4.6 eV. Therefore the lower bound of our THG’s power is estimated to be about 12 pW. The incident laser power at the sample surface is 100 mW, making the THG conversion efficiency to be in the order of $10^{-10}$ as a worst case scenario.

In terms of our detection, another limiting factor to the conversion efficiency is our inability to extend the spectral scans above 300 nm. As it can be obviously seen, we are still above the noise line at 300 nm and expect our spectral tail to go up to 310–315 nm.

Although our laser spectrum supports 5.8 fs pulses, the short wavelengths (<700 nm) do not participate in the main peak, instead they are part of the background floor of the electric field. Moreover, the components between 700 and 750nm are mostly part of the pedestals surrounding the main intensity peak. This in the end makes the excitation of the hourglass antenna less effective than possible, would we be able to have those components also in the main peak perhaps the spectrum would have been even broader.

![Figure S2](image-url)

**Figure S2:** Lower bound emission spectra of THG radiation from the bare substrate (red curve) and from the plasmonic metasurface (blue curve).
Section III: Influence of waist width of hourglass-antenna on the electromagnetic field enhancement and linear transmission spectrum

In order to further confirm that the small dip at ~720 nm in transmission arises from the enhanced absorption by the hourglass-shaped antenna, we also calculate the transmission spectrum by increasing the waist width from 30 nm to 100 nm. As shown in Fig. S3(a), the transmission dip at ~720 nm almost disappears. Meanwhile, the electric field strength at the air-gold interface [see Fig. S3(b)] and surrounding the hourglass antenna [see Fig. S3(c)] is much weaker than that one of a waist width of 30 nm. If the waist width is very narrow (such as the 30 nm in our experiments), free charges are reinforced into a very small volume, which can significantly enhance the surface current and thus optical absorption.

Figure S3: (a) FDTD-based simulation of the linear transmission spectrum of a metasurface, in which the hourglass-antenna has a 100-nm-waist width. Electric near-field maps of such an antenna structure at air-gold interface (b) and in x-z plane along the waist of the hourglass-antenna. The excitation source is a monochromatic plane wave at 720 nm, which is polarized along the y-direction and illuminates the sample from substrate to gold-film.

Section IV: Influence of bowtie on the magnetic field enhancement surrounding the hourglass-antenna

We find that the bowties can evidently influence the hourglass-antenna in terms of the magnetic field enhancement. Figure S4(a) and S4(b) artistically illustrate two neighbouring elliptical nanoapertures with and without bowties on their short axis, respectively. As shown in Fig. S4(c), compared to the magnetic field enhancement induced by pure nanoapertures (red curve), the bowties (blue curve) further enhance the magnetic field in the visible spectral region, while reducing the enhancement factor in the near-infrared region. In order to better understand the impact the bowties have on currents induced through the hourglass-antenna, Fig. S4(d-f) shows the current density distribution at three different wavelengths, as indicated
by the crosses in Fig. S4(c). At 720 nm, where the magnetic field enhancement is maximum, we find that the current densities at the bowties and at the hourglass-antenna are oscillating in-phase [see Fig.S4(d)]. There is a positive reinforcement of the current through the hourglass neck by the enhanced field at the flanks of the bowties. In contrast, at longer wavelengths [see Fig. S4(f)], the surface current at the bowties and at the hourglass display an anti-phase oscillation. Consequently, the free charges flow from the former into the latter, weakening the surface current density at the hourglass. More obviously, at 950 nm [see Fig. S4(e)], the surface current at the hourglass is completely cancelled out by the bowties, thus the magnetic field reaches a minimum at this wavelength [see Fig. S4(c)]. This further enhancement of the magnetic field via the dynamic coupling between the bowties and the hourglass boosts the THG enhancement and harmonic spectral broadening towards short wavelengths.

**Figure S4**: artistic illustration of two neighbouring elliptical nanoapertures with (a) and without (b) bowties on their short axis. (c) Calculated enhancement of magnetic field intensity with (blue curve) and without (red curve) the bowties. (d-f) Numerical simulation of surface current at 720 nm, 950 nm and 1050 nm, respectively.