Supplemental Materials

Highly efficient super-continuum generation on an epsilon-near-zero surface

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1. Dielectric function measurement

**Figure S1.** The real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) part of the epsilon for AZO thin film at 400-650 nm.

The refractive index ($n$) and extinction coefficient ($k$) of AZO films at 400-650 nm were measured by ellipsometer. Using the values of $n$ and $k$, we calculated the real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) part of the epsilon at 400-650 nm by equations:

\[ \varepsilon_1 = n^2 - k^2 \]  
\[ \varepsilon_2 = 2nk \]  

Then we fit the real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) part of the epsilon based on the Dude model:

\[ \varepsilon_1 = \varepsilon_{\infty} - \frac{\omega_p^2}{\omega^2 + \tau^2} \]
We find that the fitting results match well with the experimental results (Figure S1).

Limited by valid measuring region of ellipsometer, the epsilon of samples in near-infrared and mid-infrared regions was fitted on basis of the Dude model.

\[
\varepsilon_2 = \frac{\omega_p^2 \tau}{\omega^3 - \omega \tau^2}
\]
2. SHG spectra of AZO film under incident laser with various wavelengths

Figure S2. The SHG spectra of AZO film under incident laser with different wavelengths.
3. Characterization of third-order susceptibility by Z-scan technique

**Figure S3.** (a). The experimental setup schema of the Z-scan technique: lens (L), sample (S), iris (I) and photo detector (PD). (b). Normalized transmission (T) results of AZO achieved by the closed-aperture Z-scan technique.

The experimental setup schema of the Z-scan technique is depicted in **Figure S3 a**. In this process, a polarized Gaussian laser beam, propagating in the \( z \)-direction, is focused to a narrow waist. The sample is moved along the \( z \)-direction and the transmitted intensity is measured through a finite aperture in the far field as a function of the sample position \( z \), which is determined with respect to the focal plane.

The closed-aperture Z-scan technique was employed to characterize third-order susceptibility \( \chi^{(3)} \). The closed-aperture Z-scan measurement was performed using a femtosecond laser with the repetition frequency of 1 kHz, average power of 0.32 mW, pulse width of 100 fs, and waist spot diameter of 30 \( \mu \)m. The measured normalized transmission of AZO is shown in **Figure S3 b**. The nonlinear refractive index \( n_2 \) of AZO is fitted to be \( 4.55 \times 10^{-15} \) cm\(^2\)/W. The third-order nonlinear coefficient \( \chi^{(3)} \) can be calculated based on the nonlinear refractive index \( (n_2)^3 \) through the relation

\[
\chi^{(3)} = \frac{n \cdot n_2}{12\pi}.
\]

The obtained result of \( \chi^{(3)} = 1.42 \times 10^{-21} \) m\(^3\)/V\(^2\) at the wavelength of 1500 nm.
agrees well with the previous estimation of $4.67 \times 10^{-22} \ \text{m}^2/\text{V}^2$ from comparison with Si and previous estimation of $3.44 \times 10^{-22} \ \text{m}^2/\text{V}^2$ in theory.
4. Investigate on the influence of incident angle and laser polarization on SC spectrum

Figure S4. (a) Experimental schematic diagram of different angles between the sample and the incident laser; (b) The broadened spectrum of different laser incidence angles; (c) The experimental schematic diagram of different laser polarization directions; (d) The broadened spectrum of different laser polarization directions.

We investigated the influence of incident angle on SC spectrum. When the average laser power was 4.3 mW, by changing the angle between the sample and the laser, the SC spectra were obtained at different angles. Experimental schematic diagram was shown in Figure S4 a. It is clearly that there is no obvious change in the width of the SC spectrum as shown in Figure S4 b. According to AZO surface topography, it was clearly that the surface of AZO film was not glazed. Incident laser interacts with AZO
in all directions, so there are always electric field component in z-axis. Therefore, the coverage width of SC spectrum will not be affected by the incident angle.

To explore the influence of polarization of incident laser on the width of SC spectrum, the average laser power was limited to 4.3 mW, when the laser hit the surface of the samples. By rotating sample to change the angle between polarization direction of the incident laser and sample (Figure S4 c), we found the coverage of SC spectrum almost unchanged with the variation of the angle (Figure S4 d).
5. Fitting of supercontinuum spectra
Figure S5. (a) Computational spectrum of THG; (b) Computational Stokes Raman shift spectrum of THG; (c) Computational anti-Stokes Raman shift spectrum of THG; (d) Computational spectrum of SHG; (e) Computational Stokes Raman shift spectrum of SHG; (f) Computational anti-Stokes Raman shift spectrum of SHG; (g) Computational spectra of frequency-summing; (h) Computational spectra of four-wave mixing.

As shown in Figure 3a, the incident laser spectrum includes peak I (1247 nm-1303 nm), peak II (1303 nm-1368 nm), peak III (1368 nm-1456 nm), peak IV (1456 nm-
1564 nm), and peak V (1564 nm-1654 nm). SC spectrum was obtained by fitting incident laser with various nonlinear effect (SHG, THG, FWM and SRS. The symbol “+” and “-” represent sum and difference frequency, respectively. The symbols “Ⅰ”, “Ⅱ”, “Ⅲ”, “Ⅳ” and “Ⅴ” represent the peaks of the incident laser as shown in Figure 3a.

In Figure S5, a, b, c, d, e, f, g, h are computational spectrum of THG, computational Stokes Raman shift spectrum of THG, computational anti-Stokes Raman shift spectrum of THG, computational spectrum of SHG, computational Stokes Raman shift spectrum of SHG, computational anti-Stokes Raman shift spectrum of SHG, computational spectra of frequency-summing and computational spectra of four-wave mixing, respectively.

We can find that the calculated spectra can match the experimental spectrum very well. We also fully explain the mechanism of SC from the theory. Strong electric field of AZO surface results in giant nonlinear effect so as to induce the generation of SC spectrum.
6. The SC spectrum and the computational spectrum of SHG

As shown in Figure S6, we found that the main SHG peak shifts from 750 nm to 724 nm in the SC generation process with a frequency shift about 486 cm\(^{-1}\) by comparing SC spectrum and computational spectrum of SHG.

**Figure S6.** (a) SC spectrum and (b) the computational spectrum of SHG
7. The spectra of reflected incident laser and incident laser

Figure S7. The spectra of input laser and reflected laser

As shown in Figure S7, by comparing the spectra of reflected incident laser and incident laser, we found a little broadening in reflected incident laser spectrum with respect to the incident laser, which may owe to the strong incident laser covering up the nonlinear optical effect in the near infrared range.
8. **Study on the influence of incident power on SC spectrum**

Figure S8. The light spot photos of AZO thin film with different power of incident laser, (a) 4.3 mW, (c) 3.2 mW, (e) 2.8 mW, (g) 2.0 mW, (i) 1.3 mW; The spectra of AZO thin film with different power of incident laser, (b) 4.3 mW, (d) 3.2 mW, (f) 2.8 mW, (h) 2.0 mW, (j) 1.3 mW.

As shown in Figure S8, clearly, the intensity of light spot gradually weakens and the wavelength coverage of the spectrum also shortens with the reduction of input laser power. The detailed parameters of spectra are listed in **Table S1**.
**Table S1.** Detailed information of continuous spectra pumped by incident laser under different power intensity

| Power of incident laser (mW) | Start position (nm) | Cut-off position (nm) | Coverage (nm) |
|------------------------------|---------------------|-----------------------|---------------|
| 4.3                          | 406                 | 1100                  | 694           |
| 3.2                          | 414                 | 1100                  | 686           |
| 2.8                          | 420                 | 1100                  | 680           |
| 2.0                          | 426                 | 1100                  | 674           |
| 1.3                          | 440                 | 1100                  | 660           |

As listed in **Table S1**, the broadened spectrum coverage wavelength gradually enlarges with the improvement of input power.
References

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