Supporting Information

Ultralow Threshold Six-photon Excited Upconversion Lasing in Plasmonic Microcavity

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1. Microwire fabrication and characterization

The gain media microwires (MW) are grown using the vapor-liquid-solid (VLS) growth method (**Fig. S1**).[1] In experiment, a quartz boat with zinc powder (Alfa Aesar, 99% purity) is used as the source, and a gold film served as the catalyst for the growth of ZnO microwires. During the CVD growth process, the mixed gas of Ar (210 sccm) and O_2 (55 sccm) is injected into the furnace at 960 °C.

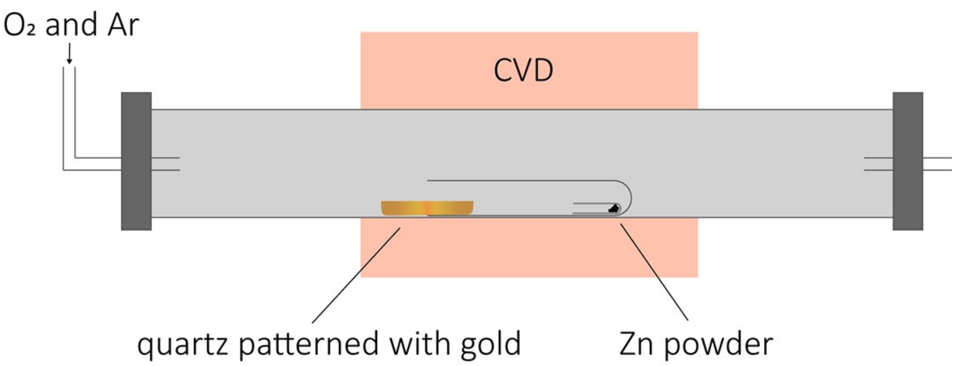

Figure S1. The sketch of chemical vapor deposition (CVD) equipment for growth of ZnO microwires.

The whole six-photon excited upconversion photoluminescence (PL) spectra of single MW is explored detail (**Fig. S2a**). Noticeable, the co-existence of upconversion fluorescence and the 3- and 5-harmonic generation of pumping light confirm the near bandedge emission (NBE) is originated from the six-photon absorption upconversion transition process (the inset of **Fig. S2a**).[2,3]

At low excitation intensity (<200 GW cm^{-2}), a broad spontaneous emission (SPE) band with linewidth of 15 nm dominates in spectrum. With increasing of input power, the NBE collapses into a spike and its intensity rises drastically. Considering the large size of MW, above superlinear behavior of the narrow spike is resulted from the multimode lasing.

To reveal the upconversion lasing (UPL) feature, the peak intensity and full-width at half maximum (FWHM) versus excitation power are given (**Fig. S2b**). The light-out via light-in (L-L) curve exhibits a well-known S-like behavior, which represents the transition from spontaneous emission to lasing oscillation in the MW cavity. The lasing threshold (P_{th}) is extracted to be 200 GW cm^{-2} from the L-L superlinear evolution. Meanwhile, the FWHM decreases to 0.2 nm as the excitation intensity raises to the threshold. Noticeably, the fitting of L-L curve (below P_{th}) using the equation $I \propto P^n$ shows the value of n-factor to be about 5.4, which is consistent with the classical nonlinear optics theory ($I \propto P^6$).[4] In addition, the solid blue line is the best fitting using the rate-equation model.

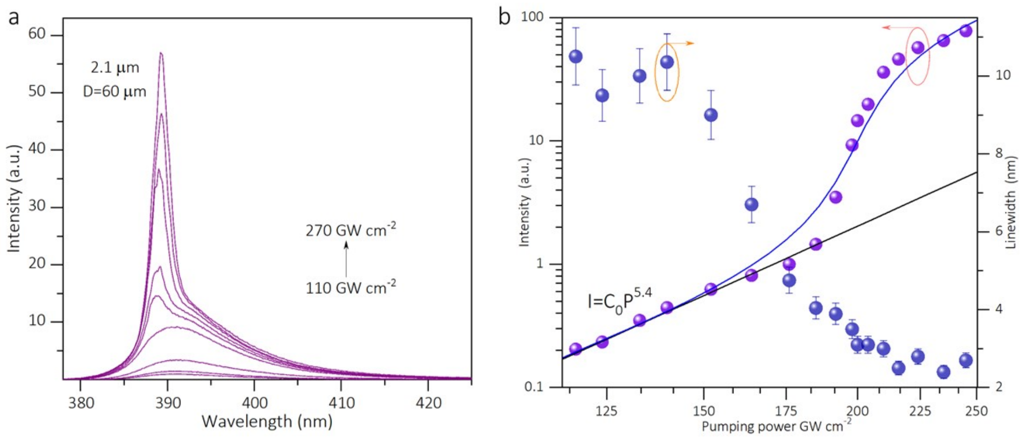

Figure S2. Upconversion lasing through six-photon excitation. a, 6-photon excited WGM lasing from bare-MW (diameter of 60 μm). Inset, the co-existence of upconversion fluorescence and the 3- and 5-harmonic generation of pumping light. **b,**

Plot of Light-out versus Light-in (L-L) and FWHM of lasing peak versus the excitation intensity. At kink point, the light is transiting from SPE to UPL emission. The solid line is the best fitting using the rate-equation model and nonlinear optical theory respectively.

In contrast to bare-MW, the polarization excitation characteristics of UPL in MW/Au-coating structure present distinct performance due to the plasmonic resonance. The variation of UPL signal with the incident light polarization angle (θ) are measured for two system, respectively (**Fig. S3**). The angle θ is defined as given in the schematic diagram (inset of **Fig. S3**). As the θ is 90 (270) degrees, UPL emission intensity are maximum in both configurations. However, the maximum UPL emission intensity of plasmonic cavity is about two times larger than bare-MW. In addition, the angle distribution of multiphoton polarization excitation in plasmonic resonator is smaller than bare-MW. Above results can be attributed to the coupled metal surface plasmonic polariton effect.[6]

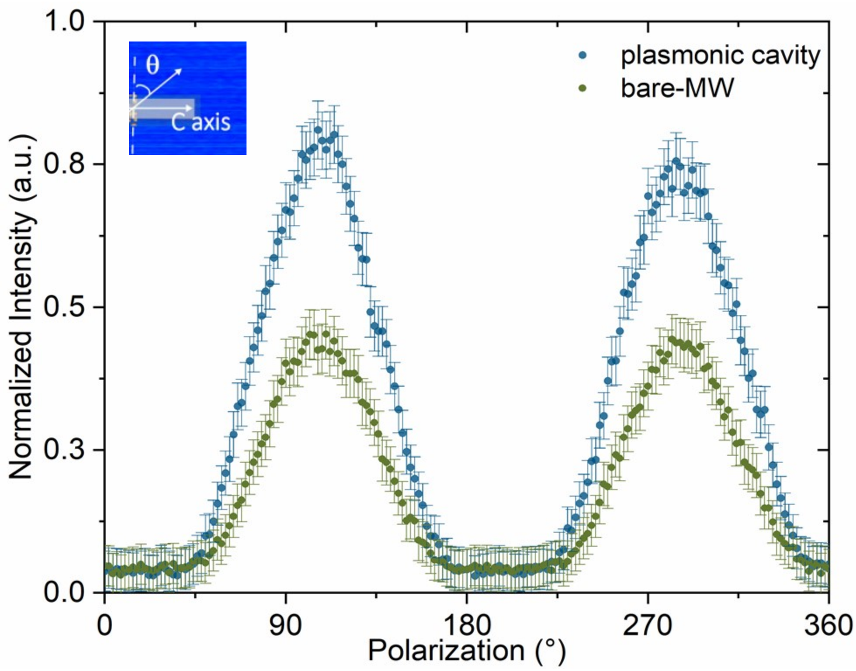

Figure S3. a, The variation of polarization excitation (1.86 μm) UPL emission for the bare-MW. Here, the angle θ is defined in the schematic diagram as shown as inset. **b**, Dependence of the polarization excitation UPL for the MW/Au-coating hybrid structure.

Figure S4 presents the distribution of field-mode in bare-MW hexagonal cavity that simulated by FDTD method. The images display that partial of optical fields are confined in the cavity. However, a portion of field-mode can leak out into free space nearby the boundary.

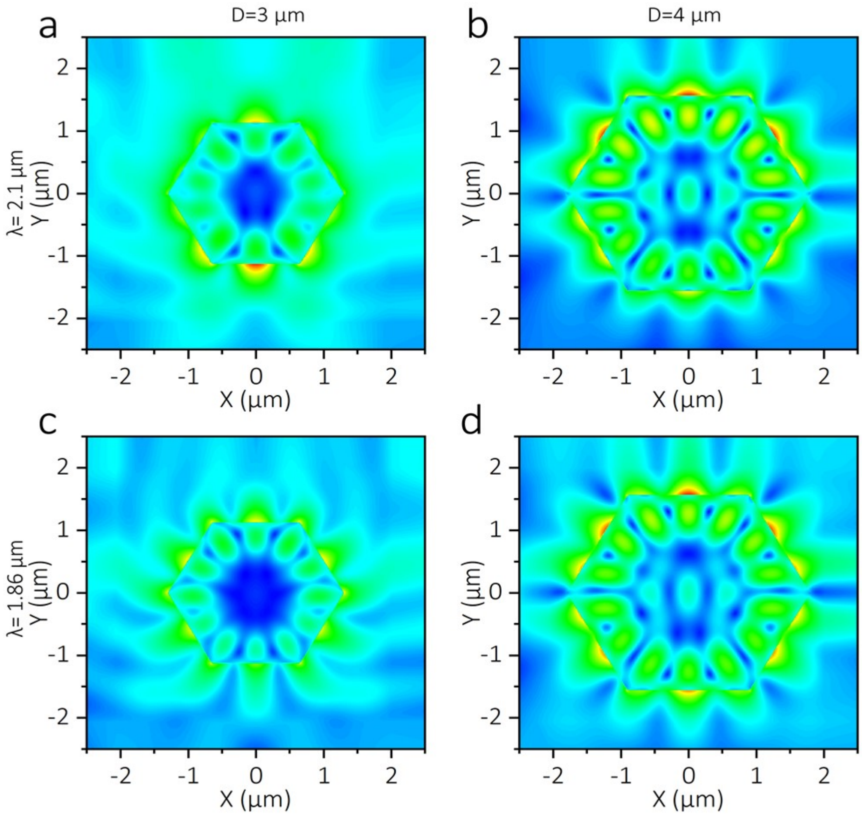

Figure S4. The field-mode in bare-MW hexagonal cavity simulated by FDTD method. a-b. The cross-sectional light field distribution of pumping light (2.2 μm) in the bare-hexagonal cavity with different diameter. **c-d.** The simulated distribution images of |E| (1.86 μm) for different cavity scale, which display the partial field-mode is confined in cavity but a little of light-field leak out from the cavity into free space.

References

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