Supporting Information

3D Zig-Zag Nanogaps Based on Nanoskiving for Plasmonic

Nanofocusing

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Figure S1. Fabrication schematic of the V-grooved silicon template. (a) $2-\mu$ m-thick photoresist was spin-coated on the silicon substrate. (b) Photoresist stripes were patterned by conventional photolithography techniques. (c) Chromium stripes were created by depositing chromium on the photoresist stripes and removing the photoresist. (d) Anisotropic KOH wet etching of the silicon substrate created the V-groove. (e) Removing the chrome one obtains the V-grooved silicon substrate.



Figure S2. FDTD simulations of the electric-field $|E|^2$ distribution in a cross-section (x × z) of a linear nanogap illuminated by non-polarized light.



Figure S3. Maximum of the calculated electric-field $|E|^2$ intensity as a function of tip angle on zig-zag nanogap structures illuminated by non-polarized light.



Figure S4. FDTD simulations the electric-field $|E|^2$ distribution of 70.5° tippednanogap directly contanted with the bulk Si substrate illuminated by non-polarized light.



Figure S5. FDTD simulations of the electric-field $|E|^2$ distribution on 70.5° tippednanogap illuminated by polarized light.



Figure S6. (a-c) FDTD simulations of the electric-field $|E|^2$ distribution on an zig-zag nanowire with tip-angles of 180°, 125.2°, 70.5° respectively, illuminated by non-polarized light. (d) Maximum of the calculated electric-field $|E|^2$ intensity as a function of tip angle on a zig-zag nanowire.



Figure S7. FDTD simulations of the electric-field $|E|^2$ distribution in nanocube with the same structural parameters (X-axis width is 100 nm, Y-axis width is 100 nm, the Z-axis height is 150 nm, the gap-width is 2.5 nm) illuminated by non-polarized light.



Figure S8. Detailed Raman spectra of the zig-zag nanogaps. Many low-intensity bands at 390, 456, 645, 783, 1003, 1140, 1179, 1390, 1438 cm⁻¹ can be observed for the 70.5° tip angle nanogap.



Figure S9. Experimentally recorded dependence of the SERS intensity versus tipangle from 70.5° to 180° in zig-zag nanowire.



Figure S10. (a, b) Schematics of the altered section directions in skiving of the Vgrooved sandwich films. (c) SEM image of the equidistant nanogap array with section direction parallel to the substrate. (d) SEM image of the nanogap array varying the spacing with slightly tilted section direction. Scale bare: $10 \ \mu m$.