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Supporting Information

Rational fabrication of silver-coated AFM TERS tips with a high enhancement and long lifetime

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The parameters in the calculation model (Fig. S1) are set according to the experimental condition, where the thickness of gold film is 10 nm and the distance between the tip and substrate is 2 nm. An oil-immersed objective with NA 1.4 was used for inverted illumination in TERS experiments. Therefore, the illumination angle θ with respect to the axis of the tip covers the range from 0 to about 70 degrees. To obtain the LSPR response for different tip radius, a clear understanding of angle dependence is necessary.

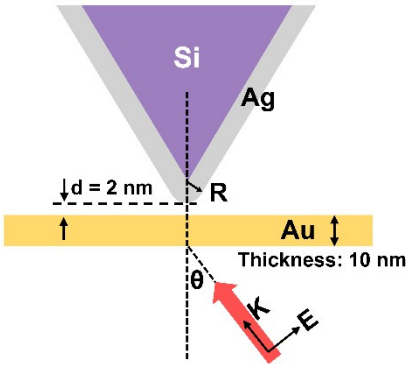

Fig. S1 Calculation model for TERS

The LSPR response of the tip and substrate system with varied illumination angle θ is calculated while the tip radius is fixed at 80 nm. Fig. S2 shows a similar LSPR response for the angles smaller than 50 degrees. When the angle is further increased to beyond the total internal reflection angle at the interface of gold film and air, the field intensity drops dramatically and renders a different electromagnetic response. Therefore, we chose 0 degree as the illumination angle to investigate the LSPR response for different tip radii, since it is able to show the typical response.

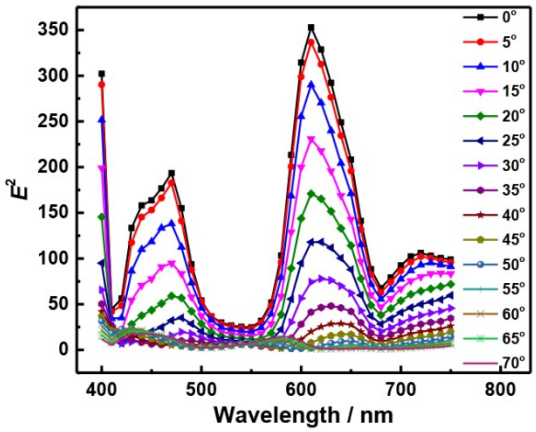

Fig. S2 Angle dependence of LSPR response for a tip radius of 80 nm

The LSPR responses for different tip radius were calculated as shown Fig. S3. According to classical understanding, Raman enhancement is contributed by the electric field intensity enhancement at both excitation and scattering wavelengths. Therefore, the field intensity enhancement factors (E^2) at the excitation wavelength (532 nm, 594 nm, and 633 nm) and at the corresponded Raman peak at 1176 cm^{-1} (577 nm, 639 nm, and 684 nm) are extracted to calculate the TERS enhancement factor at different tip radius (Fig. S4).

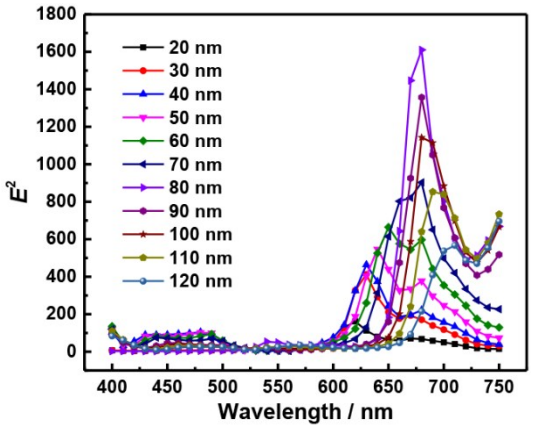

Fig. S3 LSPR responses for different tip radius illuminated along the tip axis

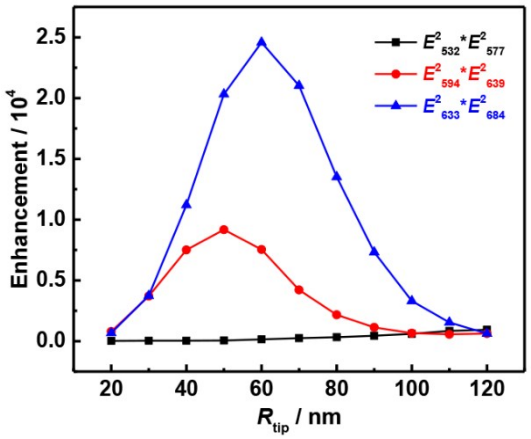

Fig. S4 The dependence of TERS enhancement on the tip radius at three wavelengths of 532, 594, and 633 nm.