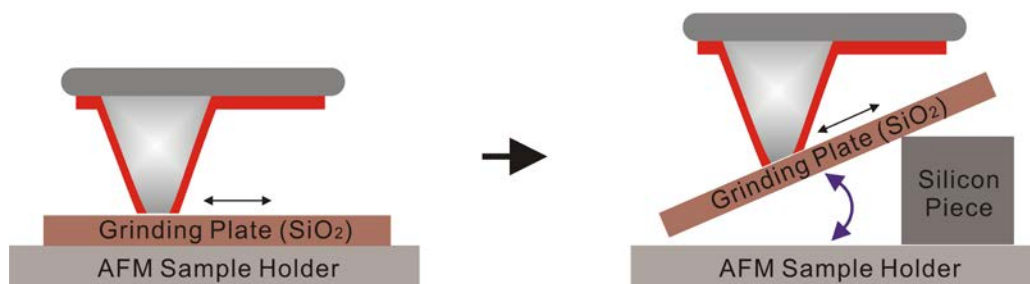


**Electronic Supplementary Information (ESI) for Analyst**

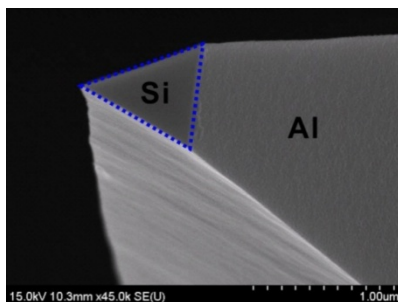
**Multilayered nano-prism vertex tip for tip-enhanced Raman spectroscopy and imaging**

Supplementary Information

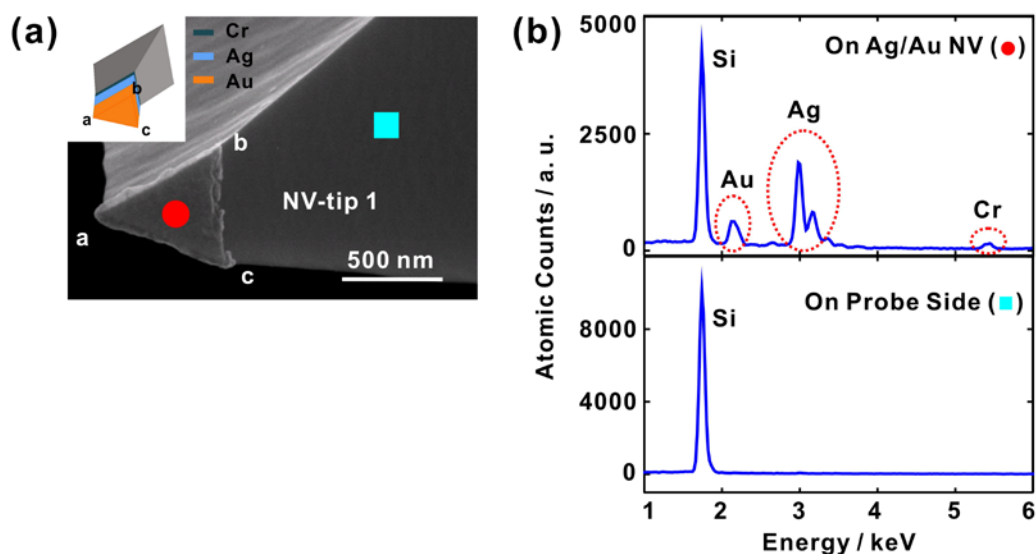
Taekyeong Kim, Ki-Seok Jeon, Kwang Heo, Hyung Min Kim, Juhun Park, Yung Doug Suh\* and Seunghun Hong\*



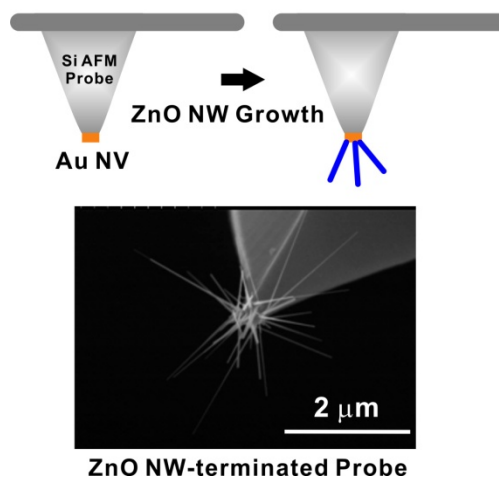
**Fig. S1** Schematic diagram depicting the grinding process of Al coated AFM tip. The grinding substrate was tilted by placing a piece of silicon under one side of the substrate, and the tip was approached onto the angled substrate for scanning.



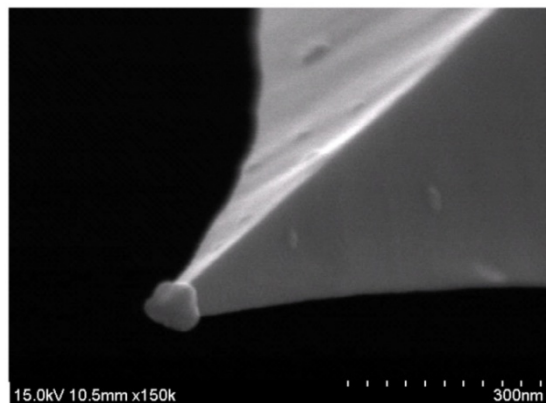
**Fig. S2** SEM image of an Al-coated silicon probe with its end ground off. The shape of the NV-tip was determined by the cross-section of the AFM probe. An AFM probe with a triangular cross-section was used to prepare the NV-tip. The multilayer thin films were deposited onto the probe and the sacrificial Al layer was removed by the chemical lift-off process, resulting in a probe terminated with a nano-prism of a triangular shape.



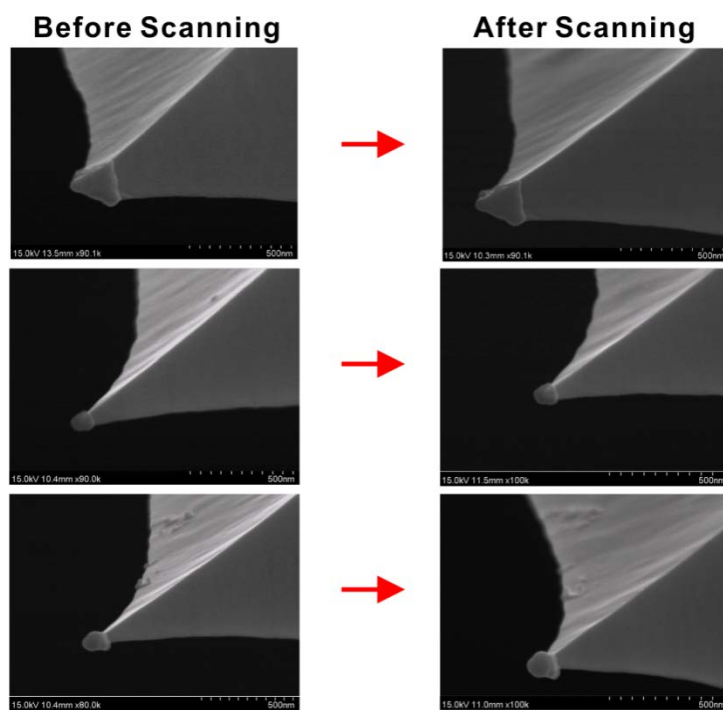
**Fig. S3** (a) SEM image of the multilayered NV tip with one triangle side of ~500 nm. (b) EDX spectrum from the Ag/Au NV (marked by a circular mark in (a)) and probe side (marked by a square mark in (a)). The signal peaks of Si, Ag, Au and Cr, were attributed to the silicon-made cantilever, Ag (~100 nm), Au (~10 nm), and Cr adhesion layer (~5 nm), respectively. In contrast, the EDX spectrum from the side of the probe showed no indication of Ag, Au, Cr or Al, implying that our process allowed us to fabricate desired nanostructures only at the end of the probe with a high definition.



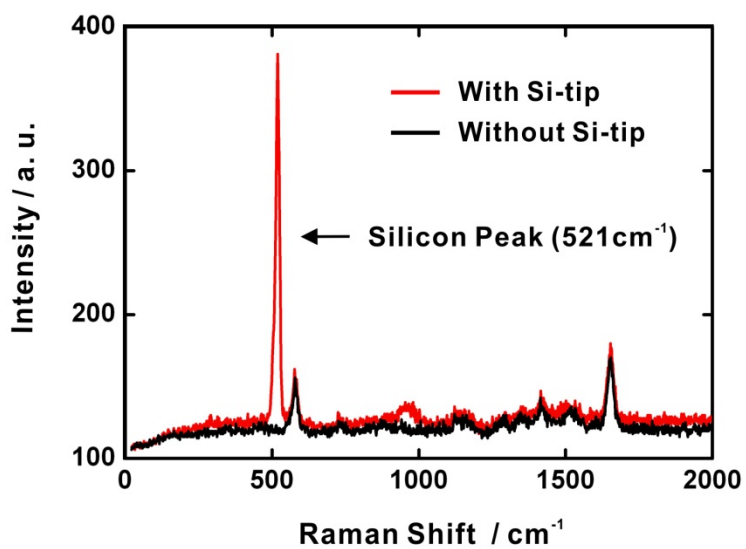
**Fig. S4** The Au nano-prism vertex (NV) was used as catalysts for the growth of ZnO NW in a chemical vapor deposition (CVD) process. The ZnO NWs could be grown only at the Au NV selectively.



**Fig. S5** SEM image of a NV-tip in ~40 nm size. The grinding conditions for the 40 nm NV-tip were scan speed 2  $\mu\text{m/s}$ , contact force ~20 nN, and humidity ~30%.

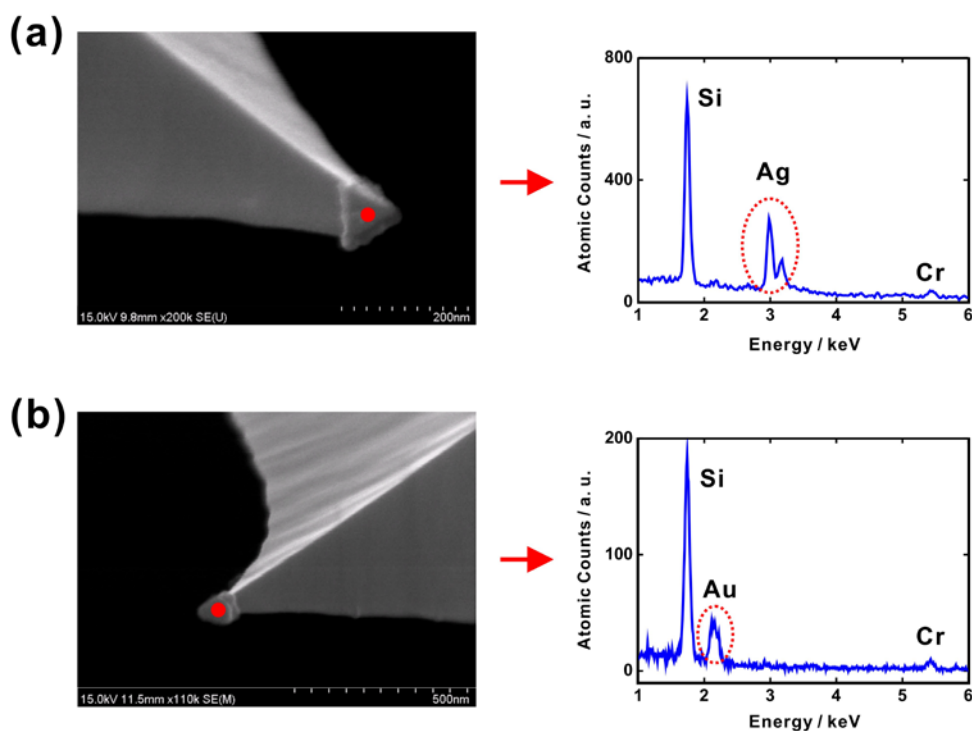


**Fig. S6** SEM images of different size NV-tips after tapping-mode AFM imaging. The NVs still remained at the end of the probe. It confirmed that our NV-tips were robust enough for stable SPM imaging.



**Fig. S7** TERS experiment on BCB molecules using normal silicon AFM tips. The signal enhancement was not observed in this case. The Si peak was observed when the Si tip was approached to the sample.





**Fig. S8** Single layer NV-tip. (a) SEM image of a Ag NV-tip and EDX spectrum on the Ag NV (marked by a red spot in the SEM image). (b) SEM image of a Au NV-tip and EDX spectrum on the Au NV (marked by a red spot in the SEM image). The “Si” and “Cr” peaks represent the silicon-made cantilever and Cr adhesion layer, respectively.