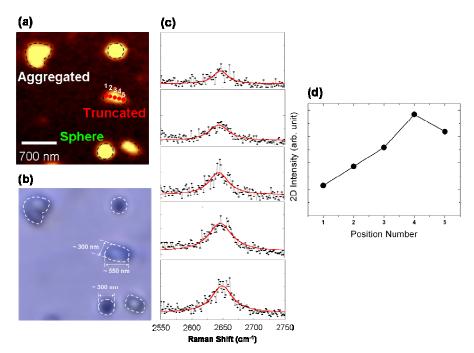
## **Supporting Information**

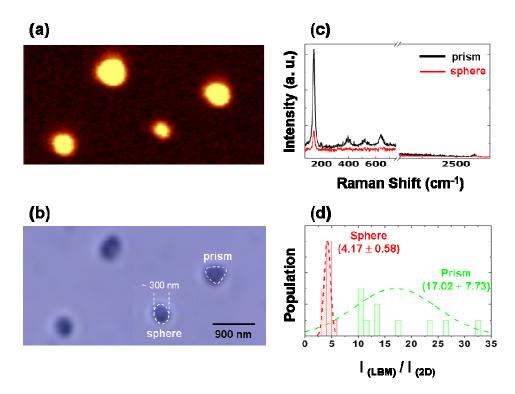
Quantification of the Relative z-polarized Electromagnetic Field Contribution and Associated Investigation of Asymmetric Shape of Layer Breathing Mode from Au Nanoparticle-Graphene-Au Film Junctions

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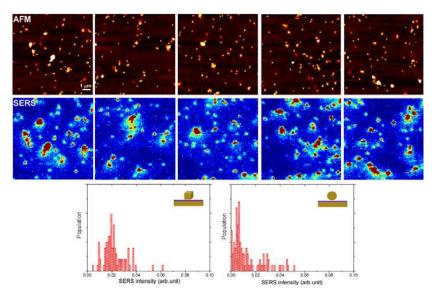
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**Figure S1.** (a) Graphene SERS image gated at LBM and (b) the corresponded optical image. The numbers from #1 to #5 on the truncated Au NP are the points where the 2D peaks are acquired and analyzed. (c) and (d) show the corresponding 2D peaks from #1 to #5 and (d) displays the 2D intensity profiles from #1 to #5 on the truncated Au NP.



**Figure S2.** (a) Graphene SERS image gated at LBM and (b) the corresponded optical image. The sphere and prism shaped Au NPs are distinguished in (b). (c) shows the representative SERS spectra at Au prism-graphene-Au TF (black) and Au sphere-graphene-Au TF. (red) (d) exhibits that Au prism case shows stronger z-polarized EM field contribution via comparing the  $I_{(LBM)}/I_{(2D)}$  values.



**Figure S3.** (up) Correlated AFM topography and SERS images of Au nanocube-Benzenethiol-Au thin film junctions. (bottom) Statistical distribution of Au nanoparticle shape dependence on SERS intensity between cube and sphere.

Reference in manuscript (complete author list)

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