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Supporting Material

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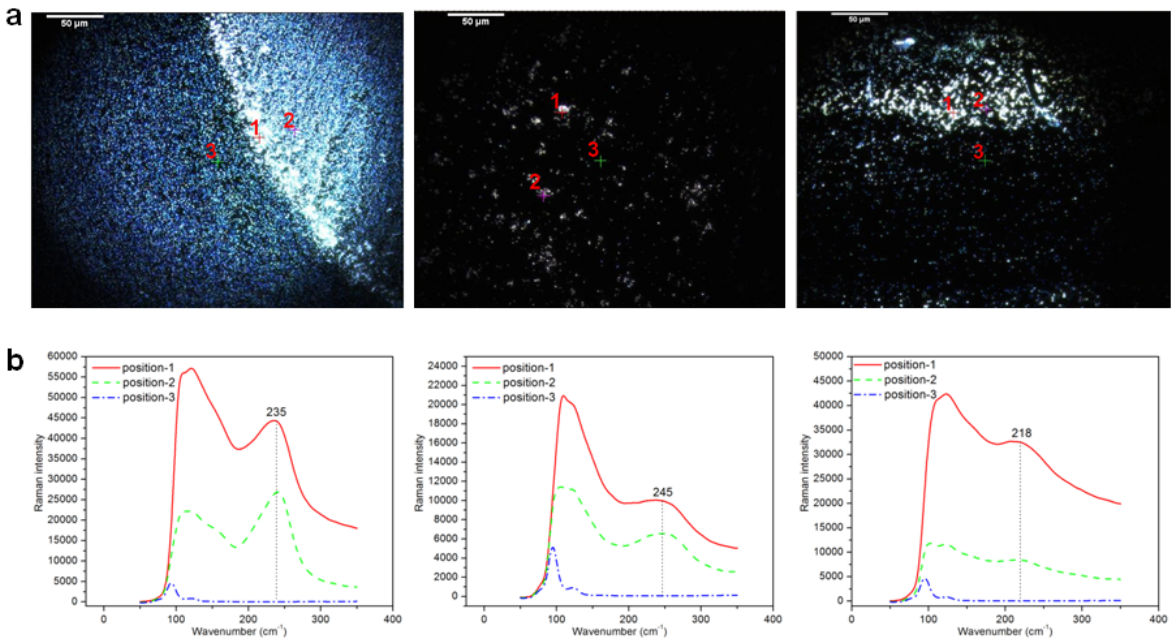

Figure S1 The plasmon-phonon mode of different SERS substrates. (a) Darkfield images of Ag nanoparticles, the Au nanoparticle film, and the Ag nanoparticle film (from left to right). Three locations were marked in each image with decreasing particle densities (position 1 to 3: dense particles, few particles, least number of particles or possible no particles). (*b*) Raman spectra of position 1 to 3 for Ag nanoparticles, the Au nanoparticle film, and the Ag nanoparticle film (from left to right).

In Fig. S1, three measurement locations with varying brightness indicating different particle densities (position 1 to 3: dense particles, few particles, least number of particles or possibly no particles) were marked in the darkfield images for Ag nanoparticles, the Au nanoparticle film and the Ag nanoparticle film (from left to right) respectively. The corresponding Raman spectra (Fig. S1 *b*) show that the intensity of the plasmon-phonon band was the highest at position-1 where particle density was the highest, and the intensity decreased at position-2 and disappeared

at position-3 where there was least number of particles or possibly no particles. These results reinforce our hypothesis that the plasmon-phonon mode inherent to metallic structures has an increasing Raman intensity to correspond with increasing surface enhancement.