Surface-enhanced Raman spectroscopy (SERS) is a powerful analytical tool that can identify chemical species at trace levels thanks to the so-called localized surface plasmon resonance. In this dissertation, four plasmonic Au nanostructures, namely, faceted ZnO/Au nanonecklace arrays, Au nanoisland arrays, nanoporous Si/Au composites, and plasma-coated SERS-active nanostructures, will be discussed with regards to their fabrication, characterization, optimization, and evaluation for SERS applications. In the end, a summary on these four plasmonic Au nanostructures will be reviewed against the standards of high-quality SERS substrates, and corresponding recommendations will be proposed to further improve the SERS performance.