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Two-Photon Vibrational Spectroscopy using local optical fields of gold and silver nanostructures KATRIN KNEIPP, Harvard Medical School, Wellman Center for Photomedicine, JANINA KNEIPP, Federal Institute for Materials Research and Testing, Berlin, HARALD KNEIPP, Harvard Medical School, Wellman Center for Photomedicine, HARVARD MIT HST TEAM, FEDERAL INSTITUTE FOR MATERIALS RESEARCH AND TESTING, D-12489 BERLIN TEAM, HARVARD MEDICAL SCHOOL, WELLMAN CENTER FOR PHOTOMEDICINE, BOSTON, MA TEAM — Spectroscopic effects can be strongly affected when they take place in the immediate vicinity of metal nanostructures due to coupling to surface plasmons. We introduce a new approach that suggests highly efficient two-photon labels as well as two-photon vibrational spectroscopy for non-destructive chemical probing. The underlying spectroscopic effect is the incoherent inelastic scattering of two photons on the vibrational quantum states performed in the enhanced local optical fields of gold nanoparticles, surface enhanced hyper Raman scattering (SEHRS). We infer effective two-photon cross sections for SEHRS on the order of 10^5 GM, similar or higher than the best known cross sections for two-photon fluorescence. SEHRS combines the advantages of two-photon spectroscopy with the structural information of vibrational spectroscopy, and the high sensitivity and nanometer-scale local confinement of plasmonics-based spectroscopy.

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