Raman effect, based on inelastic light scattering, reveals the vibrational fingerprint of molecules. However, Raman signals are much weaker than Rayleigh signals and therefore more difficult to detect. Nevertheless, an offshoot of Raman spectroscopy called surface enhanced Raman spectroscopy (SERS) has the potential to increase the signal by a factor of ten or more (10^5–10^{10}). The SERS results from plasmonic and charge transfer effects. SERS can lead to near single-molecule detection under ambient circumstances and can be used in a variety of applications, including bio-sensing, pharmaceuticals, environmental monitoring, food security, forensics, and material science.

In this doctoral thesis, SERS-active substrates for trace analyte detection are designed, fabricated, and validated. Sensitive, reproducible, and inexpensive SERS-active substrates have been realized for ultra-trace detection of analyte molecules using chemically synthesized (nanoparticle-based) and physically fabricated (EBL-based) plasmonic structures. An array of gold nano-triangles, 5-fold symmetric plasmonic quasi-crystals (PlQC) were optimised and fabricated using EBL, and harmful pesticides like leuco-malachite green (LMG) (up to 10^{-10}M) and tobacco abstinence biomarker cotinine (up to 1 ng/mL) in synthetic urine and saliva, were detected in trace amounts. Furthermore, this PlQC has also been explored for the trace detection of urea (up to 1 nM), an important biomarker of chronic kidney diseases (CKD), in synthetic urine. Additionally, a cost-effective SERS substrate was developed through the combination of silver nanoparticles with reduced graphene. This substrate can detect toxic pesticides like thiram in fruits and vegetables at concentrations up to 10^{-10} M. These substrates can be used as sensor probe for bio-diagnostics, narco-analysis, environmental monitoring, forensic science and the detection of water adulteration in trace amounts.

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