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Single molecule sensitivity in near field tip enhanced Raman scattering CATALIN C. NEACSU, MARKUS B. RASCHKE, Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy — The local-field enhancement at a sharp metallic tip in combination with resonance Raman spectroscopy provides an optical scanning probe method with ultrahigh spatial resolution. Here we report on achieving sensitivity down to the single molecule level. Illuminating the apex of a Au wire tip at variable tip-sample distances down to nm proximity results in a strong field confinement and near field coupling. This provides a highly localized light source and a controlled degree of field enhancement for Raman scattering. In the tip-scattered resonance Raman response of malachite green and rhodamine 6G molecules spectral line narrowing compared to the ensemble average and spectral diffusion are seen. Temporal fluctuations of spectral position and relative peak intensities as well as transient line splitting in time series of sequentially recorded spectra are observed. The results illustrate that single molecule Raman spectroscopy can be achieved in scattering-type near-field microscopy. This approach provides the degrees of freedom necessary for a systematic investigation and understanding of the underlying mechanisms of surface-enhanced Raman spectroscopy.

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