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Probing the molecular orientation of a single conjugated polymer via nano-gap SERS

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Determining the molecular orientation at the single molecule level is of key importance for a wide range of applications ranging from molecular electronic devices to biomedical applications. In this work, plasmonic nano-gaps consisting of a silver nanoparticle coupled to an extended silver film have been fully optimized for single molecule Surface-Enhanced Raman Scattering (SERS) spectroscopy. Finite difference time domain (FDTD) calculations revealed that the enhancement in the SERS signal is mainly associated with the dipolar mode of the nano-gap and strongly affected by the particle size which was found to be in direct agreement with our SERS measurements. In the single molecule regime, the SERS signal is dependent on the molecule orientation with regards to the field in the plasmonic nanostructure. The dipole moment derivative of the various Raman modes of the analyte were determined using Density Function Theory (DFT) calculations, and by convoluting this information with the known dipole moment of the plasmonic nanogap, we were able to recover the orientation of the single molecule within the nanogap. This analysis revealed that spin-coated conjugated polymers preferentially align their molecular chains parallel to the metallic substrate.

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