Surface-enhanced Raman detection of individual single-walled carbon nanotubes and single molecules inside

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The resonant Raman spectroscopy can only detect on-resonance single-walled carbon nanotubes (SWNTs) within the laser resonance window. In principle, surface-enhanced Raman scattering (SERS) spectroscopy can expand the resonance window. However, detection of off-resonance SWNTs by SERS remains challenging due to the difficulties in locating the SWNTs exactly at the hot spots with enormous SERS enhancements. Here, we report a facile ultrasonic spray pyrolysis method to in-situ form closely spaced polyhedral gold nanocrystals (AuNCs) on SWNTs. The fact that the edges of the AuNCs attach to the SWNTs ensures the location of SWNTs at the hot spots. Consequently, we achieve enormous enhancements that enable the detection of many off-resonance SWNTs. The enormous enhancements also allow the detection of several new Raman bands of the SWNTs that have not been reported previously.

The study of single-molecule (SM) behaviour can provide fundamental physical and chemical information otherwise obscured by the ensemble averaging due to the existence of vast molecules and accessible microstates. Large efforts have been made to immobilize the single molecules in order to in-situ study the SM behaviour. Here, we used SWNTs to encapsulate and immobilize single molecules. The interior of the nanotube can serve as a nanoscale confined space with definite and uniform environments to facilitate the SM study. We utilize SERS to achieve the ultrasensitivity that can detect the SM signals and to provide the chemical specificity that can distinguish different molecular moieties via the vibrational fingerprints. We show that a single molecule located inside the SWNT and at the SERS hot spot can not only be unambiguously detected, but also provide non-blinking and stable SERS signals with little spectral wandering, whereas signal fluctuations including temporal blinking, intensity variation, and spectral wandering are widely considered as the characteristic behaviour of single molecules in the SERS community. This approach opens new possibilities toward SM chemistry and physics in nanoscale confined space.