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Single-molecule Raman mapping with sub-nm resolution[1]

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Visualizing individual molecules with chemical recognition is a longstanding target in catalysis, bio-imaging, molecular nanotechnology, and material science. Molecular vibrations provide a valuable "fingerprint" for this identification. The spectroscopy based on tip-enhanced Raman scattering (TERS) has opened a path to obtain enhanced vibrational signals thanks to the strong localized plasmonic field originated at the tip apex. However, the best spatial resolution of the TERS imaging reported to date is still limited to a few nm, obviously not adequate for resolving a single molecule chemically. Here we demonstrate unprecedented sub-molecular Raman spectroscopic mapping with spatial resolution below 1 nm, resolving even the inner structure of a single molecule and its configuration on the surface [1]. This is achieved by creating a double-resonance nonlinear process via spectral matching, particularly by matching the resonance of the nanocavity plasmon to the downward molecular vibronic transitions [2]. Such exquisite tuning capability is provided by a combination of low-temperature ultrahigh-vacuum scanning tunneling microscopy with ultrasensitive optical detection. Our nonlinear TERS technique features the use of only a continuous wave laser rather than two pulse lasers. Our finding of Raman spectromicroscopy going intra-molecular and sub-nanometer may open up a new avenue to probe surface chemical identification, optical processes and photochemistry at the single-molecule scale.

[1] R. Zhang, et al., Nature 498, 82 (2013).
[2] Z.C. Dong, et al., Nature Photonics 4, 50 (2010).

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