Single-molecule Raman mapping with sub-nm spatial resolution

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Visualizing individual molecules with chemical recognition is a dream target in catalysis, bio-science, nanotechnology, and materials 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 and confined plasmonic field in the proximity of the tip apex. In this talk, I shall demonstrate single-molecule Raman spectroscopic mapping with unprecedented spatial resolution down to about 0.5 nm, resolving even the inner structure of a single molecule and its configuration on the surface [1]. This is achieved by a delicate spectral-matching technique that invokes a double-resonance process and resultant nonlinear optical effect, thanks to the exquisite tuning capability provided by low-temperature ultrahigh-vacuum scanning tunneling microscopy. Our nonlinear TERS technique features the use of only a continuous wave laser rather than two pulse lasers. Our findings demonstrate that Raman spectromicroscopy goes intra-molecular and sub-nanometer, which opens up a new avenue to probe chemical identification, photochemistry, and even bio-imaging, all at the single-molecule scale.

[1] R. Zhang, Y. Zhang, Z. C. Dong*, S. Jiang, C. Zhang, L. G. Chen, L. Zhang, Y. Liao, J. Aizpurua, Y. Luo, J. L. Yang, J. G. Hou*, Nature **498** (2013) 82.