

Photoswitchable vibrational nanoscopy with sub-100 nm optical
resolution

Jianpeng Ao^{1‡}, Xiaofeng Fang^{2‡}, Liyang Ma¹, Zhijie Liu¹, Simin Wu¹, Changfeng
Wu², and Minbiao Ji^{1*}

¹*State Key Laboratory of Surface Physics and Department of Physics, Human Phenome
Institute, Multiscale Research Institute of Complex Systems, Academy for Engineering
and Technology, Key Laboratory of Micro and Nano Photonic Structures (Ministry of
Education), Fudan University, Shanghai 200433, China*

²*Department of Biomedical Engineering, Southern University of Science and
Technology, Shenzhen 518055, China*

[‡] *These authors contributed equally.*

^{*}Corresponding authors: minbiaoj@fudan.edu.cn

Supplemental Material

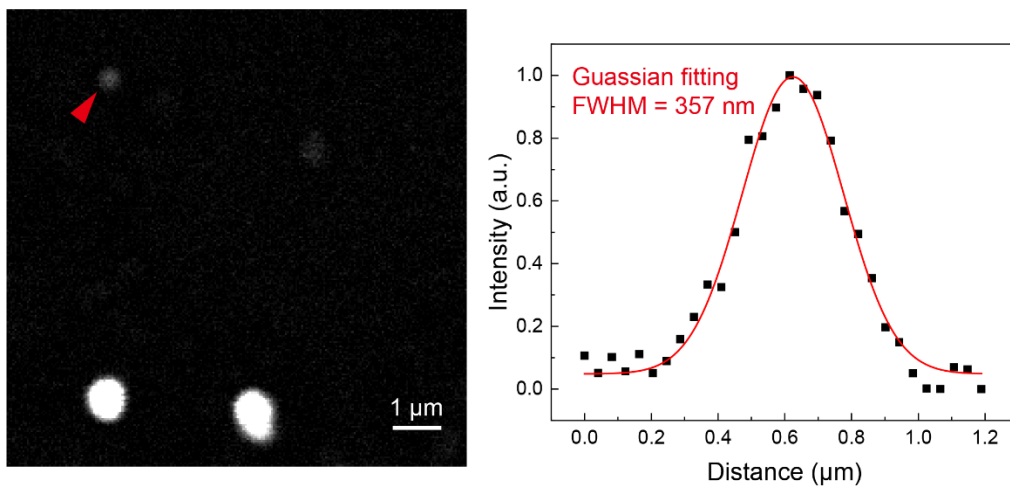


Fig. S1 PSF of SRS beams (pump and Stokes) measured with the image of gold nanoparticles.

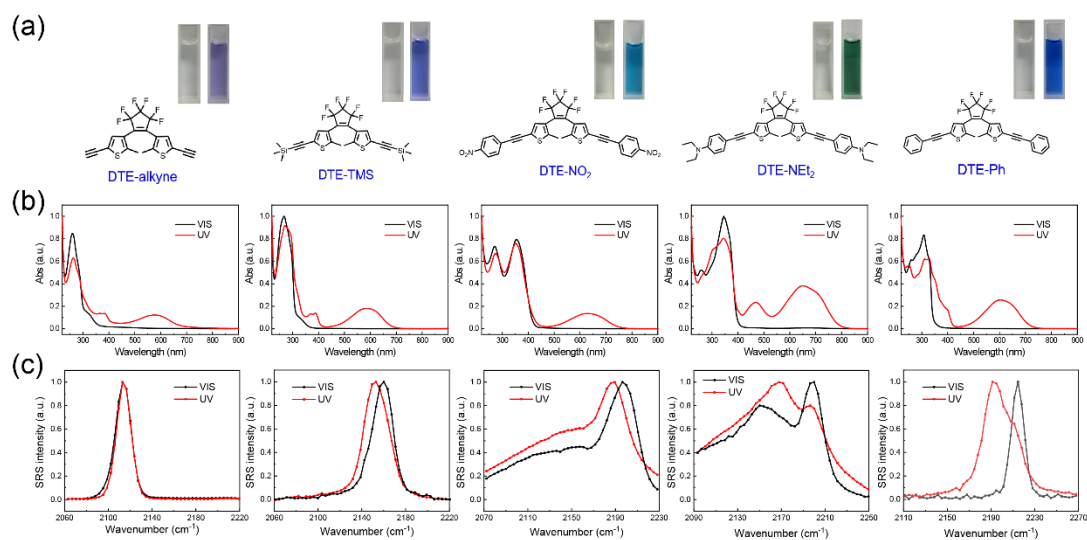


Fig. S2 Potential photochromic vibrational probes – DTE-TMS and DTE-Ph. (a) the molecular formula and the pictures of the solution; (b) the absorption spectra and (c) SRS spectra of each molecule upon visible or UV beams.

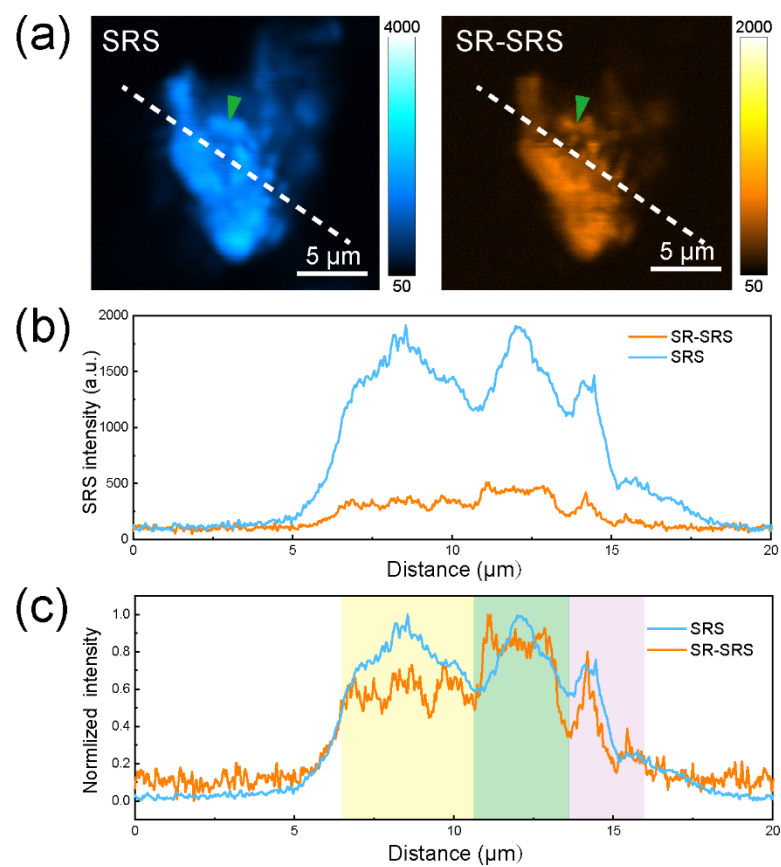


Fig. S3 Imaging on native probe molecules. (a) SRS and SR-SRS images of the probe powder; (b) raw data of the intensity profile along with the dashed lines in (a) and (c) the normalized intensity. Scale bar: 5 μm .

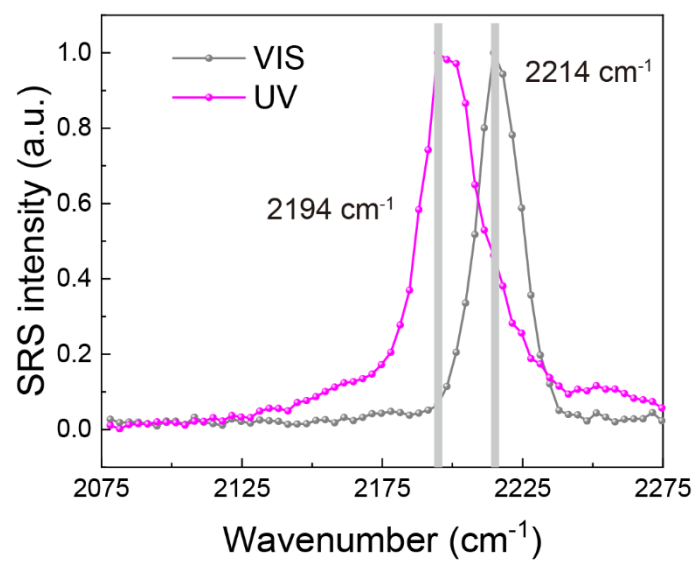


Fig. S4 The SRS spectra of DTE-Ph @ NPs upon visible or UV laser illumination.

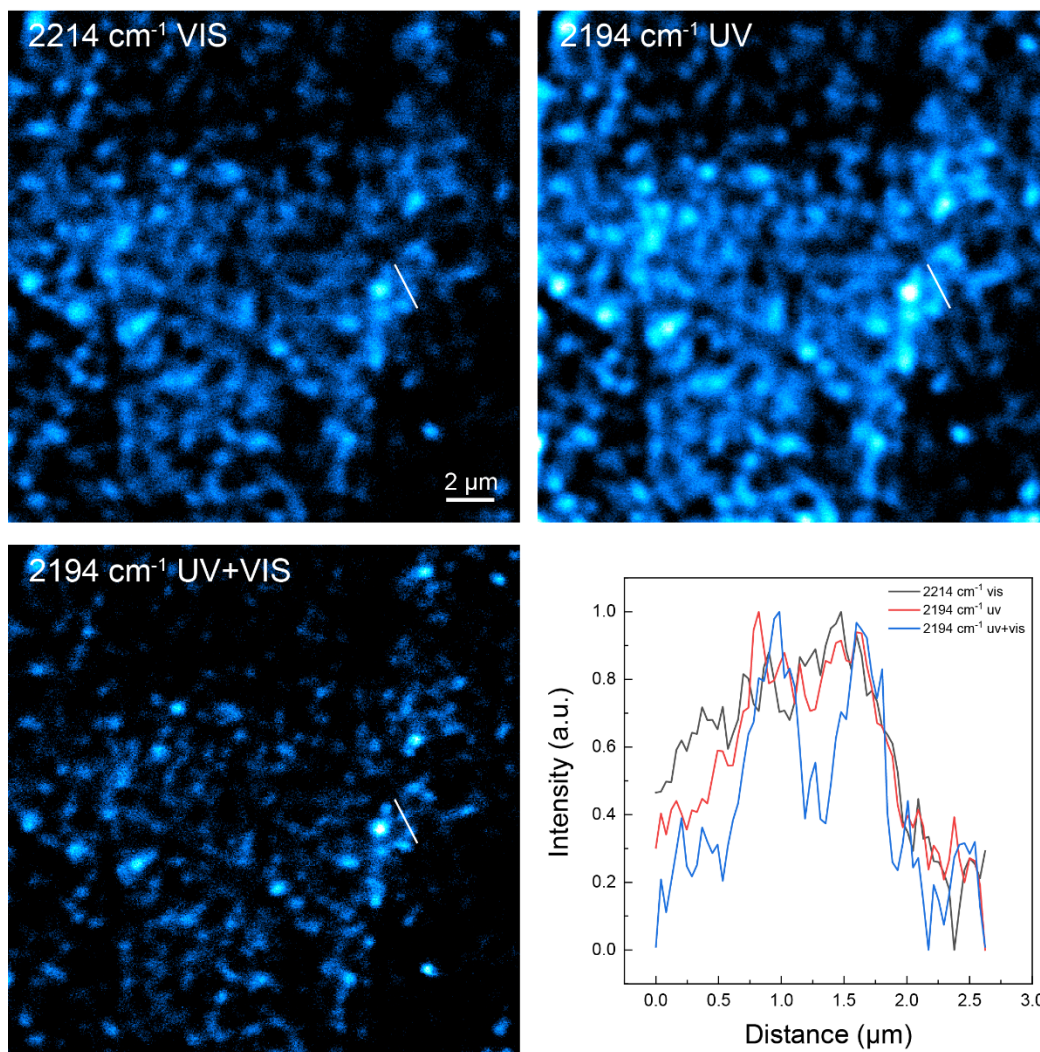


Fig. S5 SRS imaging at 2214 cm⁻¹ and 2194 cm⁻¹ coupled with different photoactive beams.

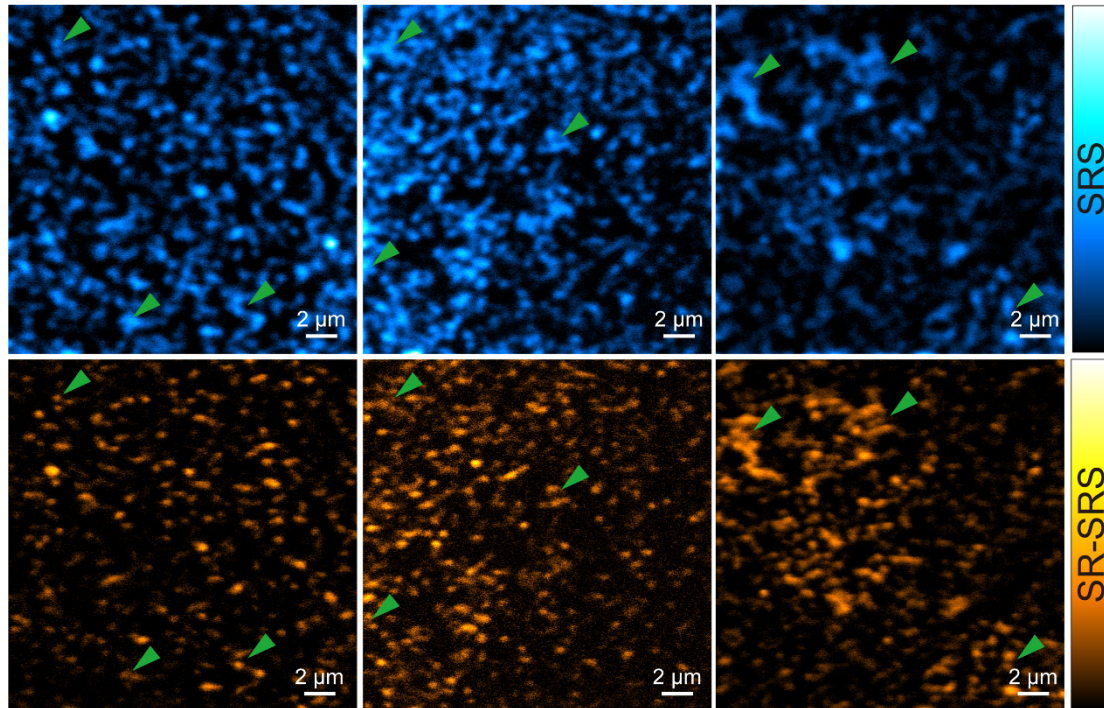


Fig. S6 Pairs of intuitive image comparison of SRS and SR-SRS of NPs and highlighted by green arrowheads. Scale bar: 2 μm .

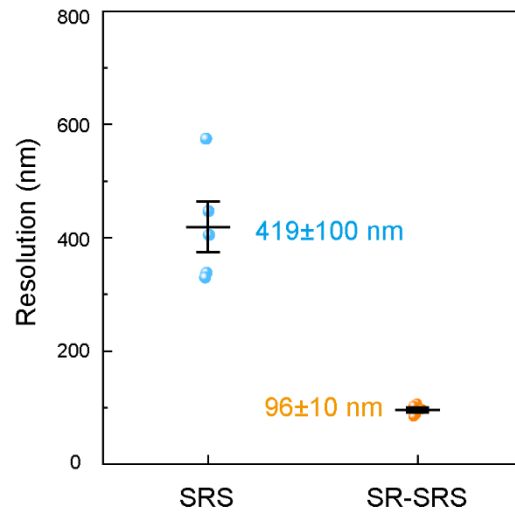


Fig. S7 Statistical analysis of multiple particles (n=5) in live cells.

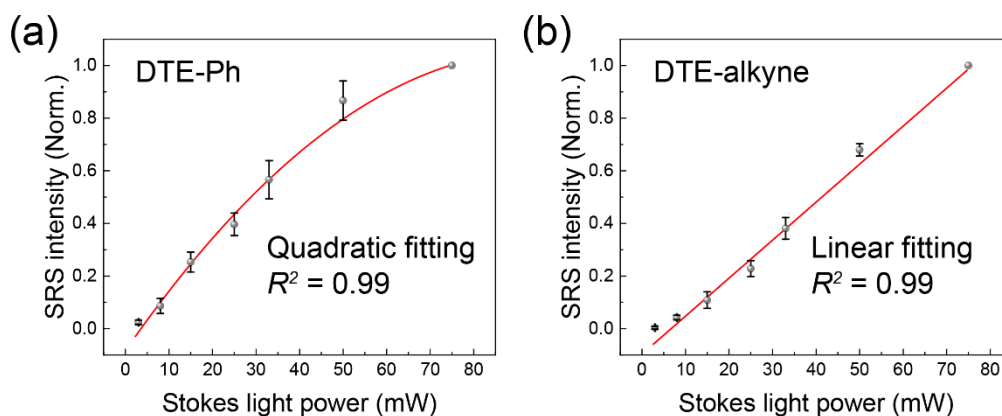


Fig. S8 Two-photon effect induced by SRS beams. (a) The relationship of SRS intensity of DTE-Ph at 2194 cm^{-1} fitted with quadratic curve and (b) DTE-alkyne at 2110 cm^{-1} fitted with linear curve with Stokes light power. DTE-alkyne is photo-inert and the SRS signal is linear with the Stokes laser power, while the SRS signal of DTE-Ph is quadratic with the Stokes laser power, indicating in high power some molecules occur ring-open reaction. ($n=3$)

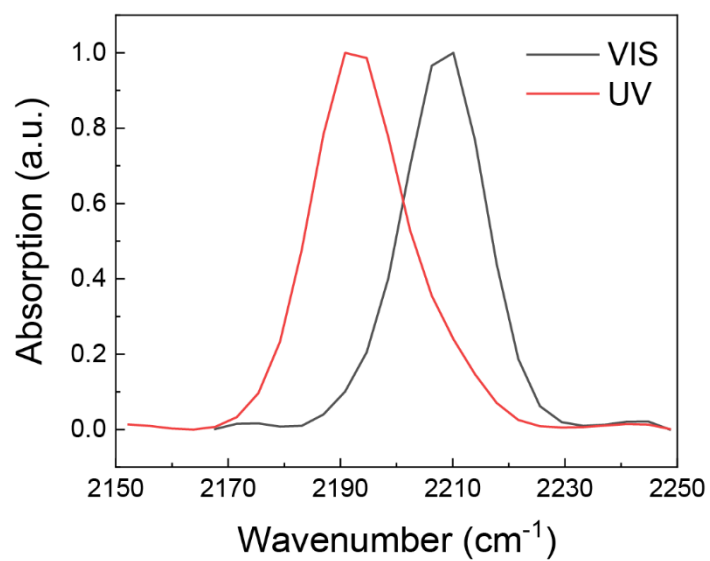


Fig. S9 Fourier transform infrared (FTIR) spectra of DTE-Ph upon visible or UV light.