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Probing single molecules with the STM in the frequency and time domains HIKARI KIMURA, Department of Physics and Astronomy, University of California, Irvine, WEICAI CAO, Department of Chemical Engineering and Materials Science, University of California, Irvine, CALVIN PATEL, Department of Physics and Astronomy, University of California, Irvine, WILSON HO, Department of Physics and Astronomy and Department of Chemistry, University of California, Irvine — We have constructed a scanning tunneling microscope (STM) and combined it with a tunable femtosecond laser (210 nm to 1040 nm) to probe single molecules with simultaneous spatial and temporal resolutions. Employing the RF lock-in amplifier to measure the laser-induced tunneling current that is directly synchronized with the high repetition rate of the laser (∼80 MHz), time resolved measurement of single molecules with atomic scale resolution can be achieved by varying the time delay between pairs of laser pulses in the two-pulse correlation or two-color pump-probe configuration. A femtosecond laser system with widely tunable wavelength enables resonant excitation of single molecules that are partially decoupled electronically from the underlying metallic substrate by a thin oxide or additionally atomic or molecular layers. The experimental arrangement allows measurement of molecular lifetimes by two-photon photoemission spectroscopy and microscopy.

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