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Ultrafast Nonlinear Optics in the Tunneling Junction

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Coupling of the electromagnetic radiation to the tip-sample junction of a scanning tunneling microscope (STM) offers exciting opportunities in molecular adsorbate identification, high-resolution dopant profiling, studies of the molecular motion and detection of dynamic changes in the electronic structure of the materials. Microwave spectral region is of particular interest because it encompasses rotational, magnetic and other resonances of molecular and solid state systems. However, previous works have either used external microwave sources or generated microwave radiation by a nonlinear mixing of the outputs from two continuous-wave lasers in a tunneling junction. In both cases, the usable spectrum was limited to a single or few frequencies. On the other hand, the regular train of pulses from a mode-locked ultrafast laser has a spectrum which represents an optical frequency comb, with a series of narrow lines (modes) spaced by the pulse repetition frequency. Here, we will show that the nonlinear response of the tunneling junction of an STM to the field of ultrashort laser pulses results in an intermode mixing that produces microwave frequency comb (MFC) with harmonics up to $n = 200$ (14.85 GHz) on both semiconducting and metallic surfaces. The observed dependence of the microwave power on the harmonic number reveals adverse effects of the tunneling gap capacitance but also shows that the roll-off at higher microwave frequencies should be negligible within the tunneling junction itself leading to intrinsic MFC spread up to THz region. We also demonstrate that MFC generation on semiconductor surface might have the same origin as THz generation in a surface depletion field. Generation of the broadband microwave signals within the tunneling junction should reduce the extraneous effects and provide significantly higher coupling efficiency. With improved frequency response, the described MFC-STM may find broad range of applications in nanoscale characterization of dynamic electronic and magnetic response of the materials in a wide frequency range.