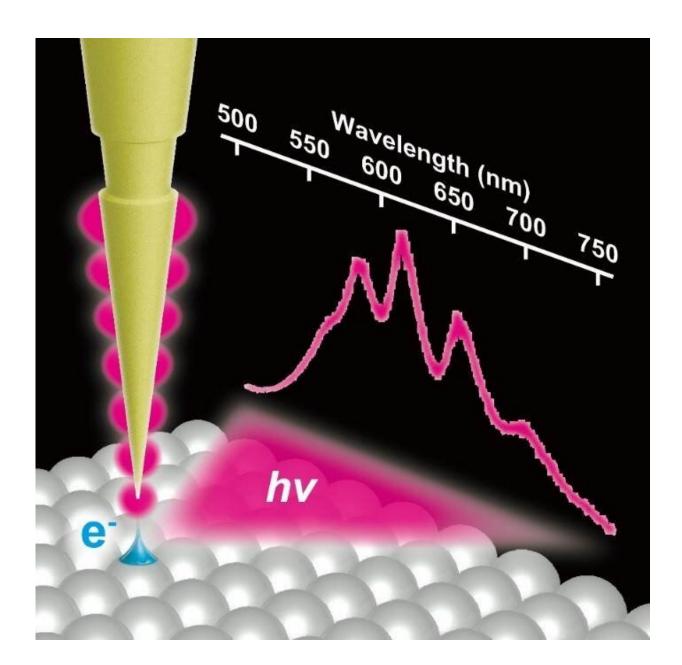


Manipulating nanoscale light in nanocavity of scanning tunneling microscope junctions

June 6 2019





Light emission from an STM junction with a plasmonic Fabry–Pérot tipNanolight (localized surface plasmon) is excited in the STM junction by tunneling electrons (e-). The emitted light (hv) shows a modulated spectrum resulting from a Fabry–Pérot interference of the propagating surface plasmon polariton on the shaft. Credit: Takashi Kumagai

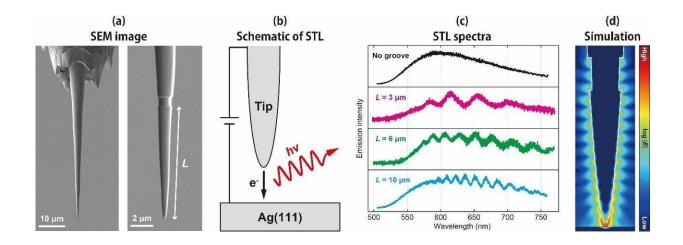
Manipulating nanoscale light in scanning tunneling microscope junctions is attained by nanofabrication of gold tips using a focused ion beam technique. Researchers at Fritz-Haber Institute, Berlin, Germany, demonstrated that a spectrum of nanolight in a nanoscale plasmonic junction can be modulated with plasmonic Fabry–Pérot tips. Precise control of nanolight is of key relevance to nanoscale imaging and spectroscopy to investigate structure, dynamics, and the optoelectronic properties of nanomaterials and single molecules.

Spatial resolution of <u>optical microscopy</u> and spectroscopy is determined by how much one can confine light in space, which is usually restricted to about half-micrometer at the best due to the diffraction limit. However, light can be confined into nanometer scale by using metallic nanostructures through excitation of localized surface plasmon <u>resonance</u> (LSPR). Having such nanolight at a sharp metallic tip is particularly useful because it can be used in scanning tunneling luminescence (STL) and scattering-type scanning near-field optical microscopy (s-SNOM) performing <u>nanoscale</u> imaging and spectroscopy to look at nanomaterials and even single molecules. However, precise manipulation of nanolight in nanoscale junction has remained an outstanding problem. Because the nature of nanolight (LSPR) is determined by the nanoscopic structure of the tip, its manipulation requires a fine processing technique at the nanoscale. In addition, nanolight confined into nanocavities is of key importance due to the strong enhancement effect of an electromagnetic field, which enables



ultrasensitive nanoscale imaging and spectroscopy.

A research team at the Fritz-Haber Institute in Berlin, headed by Dr. Takashi Kumagai, now demonstrated that manipulation of nanolight spectrum can be attained by shaping accurately plasmonic gold tips with a focused ion beam (FIB) milling technique. As an exemplary demonstration, they produced a very sharp tip with a single groove on its shaft as shown in the scanning electron microscope picture. The spectral response of nanolight confined in the nanocavity formed by the grooved tip and an atomically flat silver surface was investigated by using STL—that is, the combination of electronic and optical spectroscopies using scanning tunneling microscopy. The STML spectra with the grooved tips exhibit a characteristic modulation resulting from Fabry–Pérot type interference of surface plasmon polaritons (SPPs) on the tip shaft as the standing wave formation is visualized in the electrodynamic simulation.



Experimental results and simulation:(a) SEM images of a gold FIB-tip. A single groove is made at a distance (L) from the apex.(b) Schematic of STL measurement. Light emission (hv) occurs through excitation of LSPR in the junction by tunneling electrons (e-).(c) STL spectra obtained by FIB-tips. Spectral modulation is observed for the tips with a groove and the oscillation



period depends on the groove distance.(d) Electrodynamic simulations of twodimensional electric field mapping for the FIB-tip with a groove. Standing formation of SPP occurs on the tip shaft. Credit: Takashi Kumagai

The spectral modulation can be precisely controlled by the groove position on the shaft. They also demonstrated that the SPP Fabry–Pérot interference can be improved by optimizing the overall tip shape.

This work shows a great potential of the combination of scanning probe techniques and nano-fabrication of plasmonic tips using FIB in order to study the nature of nanolight and light-matter interactions in nanocavities, which are an important frontier of plasmonics and nanooptics. In addition, the FIB-fabricated plasmonic tips are generally applicable to s-SNOM techniques, thus paving the way for nanoscale imaging and spectroscopy with a high degree of accuracy. Moreover, spectral control of the intense near-field at the apex of <u>plasmonic</u> tips may open up new opportunities for the realization of coherent lasertriggered electron point sources for low-energy electron microscopy and holography techniques.

More information: Hannes Böckmann et al. Near-Field Manipulation in a Scanning Tunneling Microscope Junction with Plasmonic Fabry-Pérot Tips, *Nano Letters* (2019). <u>DOI: 10.1021/acs.nanolett.9b00558</u>

Provided by Japan Science and Technology Agency (JST)

Citation: Manipulating nanoscale light in nanocavity of scanning tunneling microscope junctions (2019, June 6) retrieved 26 April 2024 from <u>https://phys.org/news/2019-06-nanoscale-nanocavity-scanning-tunneling-microscope.html</u>



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