Spatial mapping of optical modes in plasmonic nanoantenna by scanning tunneling microscopy

V A Shkoldin1,2, D V Lebedev1,3, A M Mozharov1, D V Permyakov2, L N Dvoretckaia1, A O Golubok3, A A Bogdanov2, A K Samusev2, I S Mukhin1,2
1 Alferov University, 8/3 Khlopina str., St. Petersburg, 194021 Russia
2 ITMO University, 9 Kronverksky pr., St. Petersburg, 197101 Russia
3 Saint Petersburg State University, 7/9 Universitetskaya nab., St. Petersburg, 199034 Russia,
4 Institute for analytical instrumentation RAS, 26 Rizhskii pr., St. Petersburg, 190103 Russia
shkoldin@spbau.ru

Abstract. Using of inelastic electron tunnelling is very promising approach to study of subwavelength photons and plasmons sources. Such sources are very important for improving of on-chip data processing. One of the ways for development of efficient and compact optical electrically-driven sources is using of nanoantenna placed into the tunnel junction. In this work, singe optical nanoantenna was investigated under ultra-high vacuum and ambient conditions. Photon maps of nanoantenna excited under scanning tunnel microscope tip was observed and the obtained results was compared with the theoretical predictions of electromagnetic near-field distribution.

1. Introduction
In recent years, the development of electrically controlled nanoscale photon sources has become very important in the field of integrated optoelectronics. Despite numerous advantages, conventional approaches in this field, such as Fabry-Perot semiconductor lasers, solutions based on nanowires or microdisc quantum dot lasers, have resonators with dimensions significantly exceeding the operating wavelength, which limits the density of elements on a microchip. A promising approach for the implementation of nanoscale light sources is the use of a tunnel contact between two metal surfaces positioned at a sub-nanometer distance. Light emission from a metal-insulator-metal (MIM) tunnel junction was first experimentally demonstrated in the pioneering work of Lambe and McCarthy [1]. The phenomenon of photon generation was interpreted in terms of inelastic (without energy conservation) tunneling of electrons (IET), excitation of surface plasmons and their transformation into photons due to scattering by surface roughness. However, MIM type systems have one significant disadvantage, which is the extremely low efficiency of photon generation in the process of inelastic electron tunneling, namely one generated photon per 10⁴-10⁶ electrons. Thus, the task of increasing the quantum efficiency of these processes is actual.
To solve this problem plasmon nanoantenna can be used. The nanoantenna placed into tunnel junction can increase internal quantum efficient of IET up to 10% [2, 3]. The increased local density of optical states (LDOS > 10⁵) of metal nanostructure is responsible for this enhancement.

In this work the effect of a single nanoantenna on IET was investigated with scanning tunneling microscopy (STM) under ultrahigh vacuum (UHV) and ambient conditions. The used home-build STMs were coupled with optical systems to observe light emission from the tunnel junction. In addition, current-voltage measurements of tunnel junction were carried out.

2. Sample
The investigated nanoantennas were made by e-beam lithography and vacuum thermal evaporation. The gold nanoantennas had the form of cylinders and were located on the single-crystal gold film covering the mica substrate. Their height and width were equal to 50 nm and 180 nm, respectively. The crystallization of gold was made by annealing in UHV [4-6].

3. Experimental setup
The experiments were carried out with the use of two different STM setups. The first STM was UHV Omicron VT650 (Germany) with a vacuum not worse than 8.1e⁻⁹ mBar. Before the measurements STM chamber with loaded samples and tips was baked out for 8 hours at 150 °C. High quality cut Pt/Ir wires (DPT10, Bruker, USA) were used as tips. The coupled to STM optical system consisted of single photon counter IDQuantique ID120 (Switzerland) and allowed to perform STM-induced light emission mapping. The second STM was based on AIST-NT SmartSPM (USA) operating at ambient conditions. This setup allowed to collect light emission in high aperture objective, which was connected to a spectrometer Princeton Instruments SpectraPro-2500i (USA) equipped with PyLoN:400BR_eXcelon detector with liquid nitrogen cooling. For measurements at ambient conditions, we used tips made from Pt wire (300 mcm in diameter).

For UHV experiments the studied samples were loaded into the vacuum chamber and thermally annealed to remove a water film from surface. Then, STM tip was positioned under the single gold nanoantenna and spatial maps of tunnel electron-induced emission were obtained. However, the intensity of the collected light was not enough to obtain the spectrum. Additionally, current-voltage IV-measurements were carried out at numerous of points along the line crossed the nanoantenna.

To obtain STM light emission spectrum the measurements were performed at ambient conditions, where thin film of water is always present on the surface. This fact requires to use relatively high STM bias voltages (exceeding 2.5 V) in order to acquire STM-induced light maps. The STM scanning was carried out at relatively slow speed of 500 nm/sec and optical spectra was obtained with relatively short times (100 ms).

4. Experiment
At the first stage of experiments, the measurements were made in UHV STM. STM topography and maps of the emission intensity from the tunnel contact with single nanoantennas were acquired simultaneously. The results of mapping of the nanoantenna showed a nonuniform distribution of radiation pattern of the studied nanostructure. The maximum radiation intensity was observed at the edge of the disk (Figure 1a,b).

The numerical simulations show that two optical modes can be excited in the nanoantenna. With the probe positioned in the center of the nanoantenna, only one mode at 668 nm can be excited. When the STM probe is positioned at the edge of the nanoantenna, two modes can be efficiently excited at 668 nm and 566 nm (Figure 1c). Both modes have a «hot spot» at the edge of the nanoantenna, which leads to increasing LDOS at the periphery of the structure. This explains the increasing of the emission process efficiency from the tunnel contact, which was observed in the experiment (see Figure 1a, b). IV-curves analysis also shows a change of LDOS at the edge of the nanoantenna.
The optical spectra of the nanoantenna STM-induced emission contain two clear bands with maxima at 670 nm and 765 nm (Figure 2). The long-wavelength band dominates in the spectra obtained on the substrate (see green lines in Figure 2). And the short-wavelength band dominates in the spectra obtained on the nanoantenna (see blue lines in Figure 2). When the STM tip was located at the edge of nanodisk, optical spectra had the enhanced short-wavelength component. These experimental results qualitatively correspond to the results of numerical simulations, at the periphery of nanodisk the mode associated with the shorter wavelength component can be excited. We suggest that the difference of the peak positions predicted by modelling can be caused by the changing of metal work function at ambient conditions, as well as the imperfections of disk fabrication. The simulation was performed for an ideal cylinder in vacuum, while in air the sample surface is covered with a thin layer of water, which affects the parameters of the tunnel contact, including the energy position of Fermi level. The low selectivity of mode excitation is most likely related to the relatively large diameter of the STM tip, which is comparable with the size of the nanoantenna.

![Figure 1](image1.png)

**Figure 1.** (a) STM topography image and corresponding STM light emission map of a single nanodisk on the surface of a gold film. (c) Simulated spectrum of radiative power of the system for different tip positions.

![Figure 2](image2.png)

**Figure 2.** Radiation spectra of the tunnel contact obtained at different positions. The blue lines are spectra obtained from the tunnel contact with a nanoantenna. The green lines are typical spectra obtained from the gold substrate.

5. Conclusion
The single nanoantennas were investigated by STM at UHV and ambient conditions. The spatial distribution map of STM-induced light intensity was acquired. It was shown that radiation power depends on the tip position relative to the nanoantenna. The experimental results correspond well to the numerical simulations. Also, optical spectra of STM-induced light emission from gold nanoantenna were obtained, and they are governed by the optical resonances of the investigated nanostructures.
Acknowledgments
This work was done with the financial support of the Russian Federation President Council (MK-2428.2020.2) and the Russian Foundation for Basic Research (project 19-32-90028). D.V. Lebedev and A.O. Golubok thanks for financial support in the formation of gold samples the Russian Science Foundation (Project No. 21-79-10346)

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