Conference materials
UDC 535
DOI: https://doi.org/10.18721/JPM.183.218

STM visualization of local density of optical states of gold nanoantennas with sub-diffraction resolution

N.A. Solomonov[1,2✉], D.V. Lebedev[1], D.V. Pavlov[3], A.A. Kuchmizhak[3,4], A.O. Golubok[5], I.S. Mukhin[2]

[1] Alferov University, St. Petersburg, Russia;

[2] Peter the Great St. Petersburg Polytechnic University, St. Petersburg, Russia;

[3] Institute of Automation and Control Processes FEB RAS, Vladivostok, Russia;

[4] Far Eastern Federal University, Vladivostok, Russia;

[5] Institute for Analytical Instrumentation RAS, St. Petersburg, Russia

✉ solomonov.nik@gmail.com

Abstract. Simultaneous recording of luminescence induced by inelastic electron tunneling together with topography in scanning tunneling microscopy (STM-L) is a powerful tool in the study of local optical properties of nanostructures. In this study, we use the STM-L technique to visualize the local density of optical states (LDOS) by detecting the optical signal generated in the tunnel junction between a tungsten STM probe and a hollow hemispherical gold nanoantenna (d = 500 nm, h = 300 nm) fabricated by femtosecond laser printing. The light emission from the tunnel gap is caused by the process of inelastic tunneling of electrons. The intensity of photon signal directly dependent on LDOS — a key parameter in the design of light emitting inelastic tunneling-based nanoscale photon sources. We demonstrate that the STM-L method can provide information on the LDOS features of individual nanoantenna with sub-diffraction spatial resolution, as well as reveal smooth LDOS variations in periodic arrays of nanoantennas. These results highlight the unique capability of STM-L to reveal nanoscale optical patterns inaccessible to conventional diffraction-limited microscopy, highlighting its potential for advanced optical characterization at the nanoscale.

Keywords: gold nanoantenna, inelastic tunneling of electrons, local density of optical states, sub-diffraction imaging, femtosecond laser printing, scanning tunneling microscopy

Funding: The work was supported by the Russian Science Foundation (grant no. 25-19-00666).

Citation Solomonov N.A., Lebedev D.V., Pavlov D.V., Kuchmizhak A.A., Golubok A.O., Mukhin I.S., STM visualization of local density of optical states of gold nanoantennas with sub-diffraction resolution, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 18 (3.2) (2025) 96–100. DOI: https://doi.org/10.18721/JPM.183.218

Материалы конференции
УДК 535
DOI: https://doi.org/10.18721/JPM.183.218

Визуализация локальной плотности оптических состояний золотых наноантенн с субдифракционным разрешением в сканирующем туннельном микроскопе

Н.А. Соломонов [1, 2 ✉], Д.В. Лебедев [1], Д.В. Павлов [3],
А.А. Кучмижак [3, 4], А.О. Голубок [5], И.С. Мухин [2]

[1] Академический университет им. Ж.И. Алфёрова РАН, Санкт-Петербург, Россия;

[2] Санкт-Петербургский политехнический университет Петра Великого, Санкт-Петербург, Россия;

[3] Институт автоматики и процессов управления ДВО РАН, г. Владивосток, Россия;

[4] Дальневосточный федеральный университет, г. Владивосток, Россия;

[5] Институт аналитического приборостроения РАН, Санкт-Петербург, Россия

✉ solomonov.nik@gmail.com

Аннотация. Мультиканальная регистрация люминесценции, возбуждаемой зондом сканирующего туннельного микроскопа (СТМ-Л), и туннельная спектроскопия с регистрацией оптического сигнала (СТС-Л) доказали свою эффективность в качестве мощных инструментов для одновременного исследования оптических и электрических свойств источников фотонов. В данном исследовании мы используем технику СТМ-Л для визуализации локальной плотности оптических состояний (ЛПОС) путем регистрации оптического сигнала, генерируемого в туннельном переходе между вольфрамовым зондом СТМ и полой полусферической золотой наноантенной (d = 500 нм, h = 300 нм), изготовленной с помощью фемтосекундной лазерной печати. Световое излучения из туннельного зазора вызвано процессом неупругого туннелирования электронов (НТЭ), а его интенсивность напрямую зависит от ЛПОС — ключевого параметра в разработке наноразмерных источников фотонов. Мы демонстрируем, что СТМ-Л позволяет получать информацию о тонких пространственных особенностях ЛПОС как на уровне отдельных наноструктур с разрешением ниже дифракционного, так и выявлять плавные изменения ЛПОС в периодических массивах наноантенн. Эти результаты подчеркивают уникальные возможности СТМ-Л для продвинутых исследований оптических и электрических свойств наноразмерных источников фотонов, недостижимых для традиционной оптической микроскопии с дифракционным ограничением.

Ключевые слова: золотая наноантенна, неупругое туннелирование электронов, локальная плотность оптических состояний, субдифракционные оптические измерения, фемтосекундная лазерная печать, сканирующая туннельная микроскопия

Финансирование: Работа выполнена при поддержке Российского научного фонда (грант № 25-19-00666).

Ссылка при цитировании: Соломонов Н.А., Лебедев Д.В., Павлов Д.В., Кучмижак А.А., Голубок А.О., Мухин И.С. Визуализация локальной плотности оптических состояний золотых наноантенн с субдифракционным разрешением в сканирующем туннельном микроскопе // Научно-технические ведомости СПбГПУ. Физико-математические науки. 2025. Т. 18. № 3.2. С. 96–100. DOI: https://doi.org/10.18721/JPM.183.218

Introduction

The development of nanoscale light sources control by electrical signals remains a key challenge in modern optoelectronics and integrated photonics. Among the various approaches to implement such device, light emission via inelastic tunneling of electrons in metal–insulator–metal (MIM) and metal–insulator–semiconductor (MIS) tunnel junctions have attracted significant attention as a promising platform for electrically-driven photon generation at the nanoscale [1–3]. In such MIM and MIS systems, electrons can tunnel through a sub-nanometer insulating layer (tunnel gap) losing part of their energy through the excitation of various states across a broad energy spectrum. This includes electron–hole pairs, molecular vibration transitions, localized plasmon modes (gap plasmon), propagating surface plasmons, and other states near the sample surface. Some of these excitations may decay radiatively, resulting in photon emission that can be detected using an external optical system [4]. However, only a small part of the tunneling electrons participates in the inelastic process, which leads to a low external quantum efficiency of the LEIT process., This fact significantly limits the application of LEIT for nanoscale light source technologies.

One of the strategies to enhance photon yield is to introduce resonant nanostructures, such as plasmonic nanoantennas, which create a strong enhancement of the local electromagnetic field into the tunneling region [5, 6]. These structures can significantly enhance the local density of optical states (LDOS), thereby increasing the probability of photon emission during inelastic tunneling process. As a result, properly designed nanoantennas can significantly improve the radiation efficiency of tunneling transitions and facilitate the development of compact nanoscale electrically-driven light sources.

In this work, we employ multichannel registration of luminescence excited by a scanning tunnelling microscope probe (STM-L) to visualize subtle spatial features in the LDOS of hollow hemispherical gold nanoantennas fabricated via femtosecond laser printing. We observe distinct LDOS patterns at the individual nanoantenna level associated with the excitation of intrinsic plasmonic modes, as well as smoothly varying LDOS distributions across periodic antenna arrays arising from propagating surface plasmon waves and their subsequent scattering into photons within the visible spectral range.

Materials and methods

The investigated sample consisted of a 50 nm thick gold film deposited on an optically smooth glass substrate using electron beam evaporation without any adhesion layer. Hollow arrays of hemispherical gold nanoantennas with a base diameter of 500 nm and a height of 300 nm were fabricated on the gold surface via direct femtosecond laser printing [7]. The fabrication was performed using a pulsed Ti:sapphire laser system (λ = 800 nm, pulse duration ~100 fs, repetition rate of 80 MHz) focused onto the metal film through a high-numerical-aperture objective.

Tunnel junction was formed between the fabricated nanostructures and an electrochemically etched tungsten STM tip in an ultrahigh vacuum scanning tunneling microscope (Omicron VT STM/AFM). Photon emission from the tunnel gap was recorded using an IDQuantique ID120 (350–1000 nm) single-photon counting module synchronized with the acquisition of topographic data in the STM.

Results and Discussion

Fig. 1 shows the typical STM topography maps and corresponding maps of the spatial distribution of the recorded optical signal (STM-L) for a single hollow gold nanoantenna and a periodic array of nanoantennas. The STM measurements were carried out with scan resolutions of 200×200 points over a 1×1 μm area for the single nanoantenna, and 150×150 points over a 4×4 μm area for their array. The tunneling parameters were fixed at a bias voltage of 2.5 V and a tunneling current of 1 nA. All measurements were performed at room temperature under ultrahigh vacuum conditions (~10^{-10} mbar).

In the case of a single nanoantenna (Fig. 1, *a*, *b*), the photon emission observed in the STM-L map exhibits a distinct spatial pattern, with increased intensity both at the edges and center of the antenna. This reflects an inhomogeneous distribution of the local density of the nanoantenna LDOS, which can be attributed to the excitation of intrinsic plasmonic modes of nanoantenna.

For a periodic array of nanoantennas (Fig. 1, *c*, *d*), the STM-L signal shows both localized emission from individual antennas and smoothly varying background component. The gradual variation in the emission intensity, observed when the STM probe is positioned between the nanoantennas, can be attributed to the excitation of propagating surface plasmon waves launched by inelastic electron tunneling. These waves subsequently interfior and scatter by the periodic nanoantenna structure, resulting in the emission of photons in the visible frequency range, which are registered by the optical detection system.

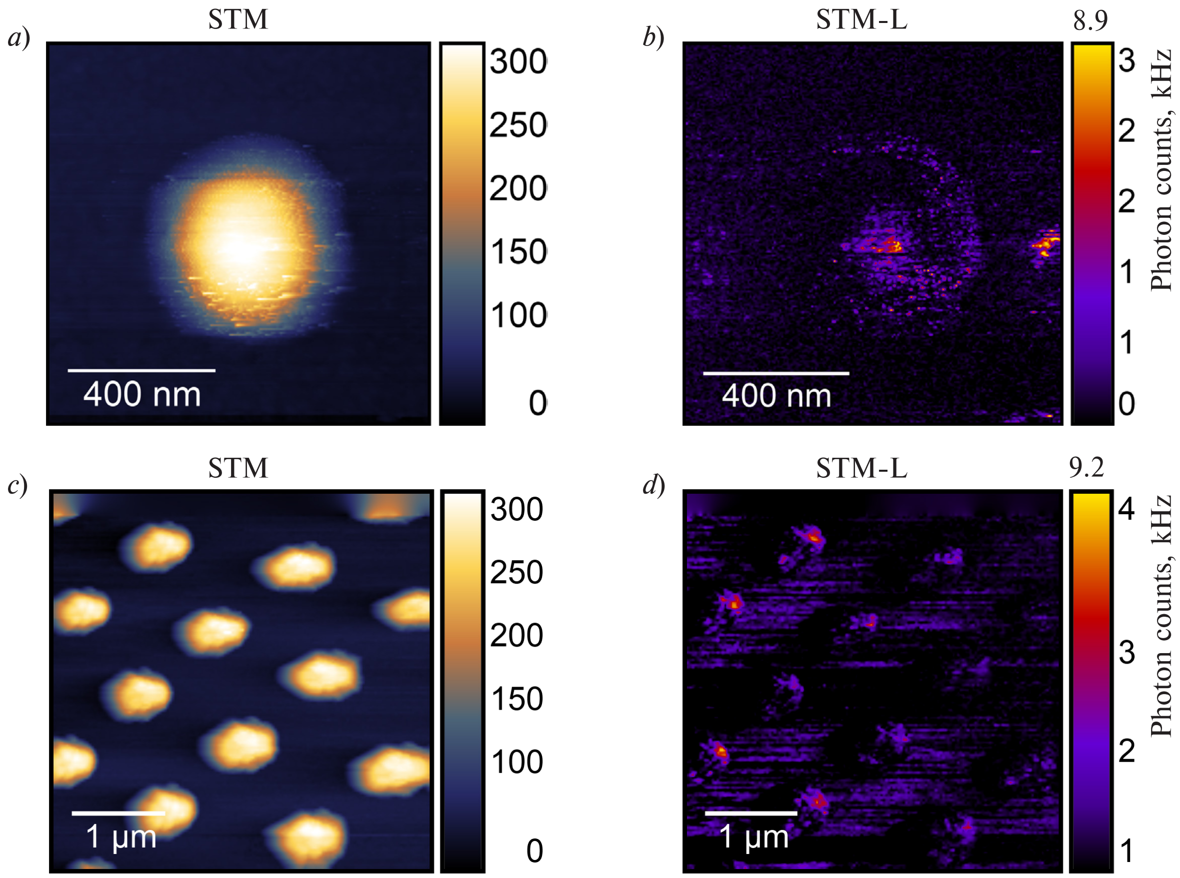

Fig. 1. STM-L measurement of the studied systems: single nanoantenna (*a*, *b*) and periodic nanoantenna array (*c*, *d*): (*a*, *c*) STM topography (*b*, *d*) spatial distribution of the recorded optical signal (STM-L)

Conclusion

This study demonstrates the potential of STM-L to resolve fine spatial variations of LDOS in plasmonic nanostructures. We have observed characteristic LDOS distributions in single hollow gold nanoantennas, including patterns corresponding to the excitation of intrinsic plasmonic modes caused by inelastic electron tunnelling. Smoothly varying intensities (or LDOS) of the recorded radiation were found in periodic arrays, explained by the excitation and scattering of propagating surface plasmon waves with emission of photons in the visible range.

The results confirm the potential of STM-L as a power tool for studying light-matter interactions with sub-diffraction resolution at the level of individual nano-objects.

REFERENCES

1. **Lebedev D.V., Solomonov N.A., et al.,** Femtosecond Laser-Printed Gold Nanoantennas for Electrically Driven and Bias-Tuned Nanoscale Light Sources Operating in Visible and Infrared Spectral Ranges, The Journal of Physical Chemistry Letters 14 (22) (2023) 5134–5140.

2. **Lebedev D.V., Solomonov N.A., et al.,** Electrically-Driven Light Source Embedded in a GaP Nanowaveguide for Visible-Range Photonics on Chip, Advanced Optical Materials. 12 (25) (2024) 2400581.

3. **Solomonov N.A., Lebedev D.V., et al.,** Luminescence enhancement in inelastic tunnelling of electrons by changing the geometry of the tunnelling contact, St. Petersburg State Polytechnical University Journal. Physics and Mathematics. 17 (3.2) (2024) 236–240.

4. **Lebedev D.V., et al.,** Indirect Detection of the Light Emission in the Local Tunnel Junction, Physica status solidi (RRL)–Rapid Research Letters. 14 (3) (2020) 1900607.

5. **Lebedev D.V., et al.,** Nanoscale electrically driven light source based on hybrid semiconductor/metal nanoantenna, The Journal of Physical Chemistry Letters. 13 (20) (2022) 4612–4620.
6. **Kern J., et al.,** Electrically driven optical antennas, Nature Photonics. 9 (9) (2015) 582–586.
7. **Pavlov D.V., et al.,** Third-Harmonic Generation in Plasmonic Metasurfaces Fabricated by Direct Femtosecond Laser Printing, JETP Letters. 119 (10) (2024) 763–767.

THE AUTHORS

SOLOMONOV Nikita A.
solomonov.nik@gmail.com
ORCID: 0000-0003-0675-8659

LEBEDEV Denis V.
denis.v.lebedev@gmail.com

PAVLOV Dmitriy V.
pavlov_dim@mail.ru

KUCHMIZHAK Aleksandr A.
ku4mijak@iacp.dvo.ru
ORCID: 0000-0002-5376-5555

GOLUBOK Alexander O.
golub@ianin.spb.su
ORCID: 0000-0001-9970-9172

MUKHIN Ivan S.
imukhin@yandex.ru

Received 02.09.2025. Approved after reviewing 02.10.2025. Accepted 02.10.2025.