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Luminescence enhancement in inelastic tunnelling of electrons by changing the geometry of the tunnelling contact

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Abstract: We have experimentally investigated the light emission resulting from inelastic electron tunneling in the transition with hemispherical gold nanoantennas (d = 400 nm, h = 300 nm) created by direct fs-laser ablation. We found two characteristic modes of luminescence: standard - increased signal is observed in the region of nanoantennas at tunneling currents below 2.25 nA and inverted-anomalous, where the gold surface is mainly luminescent, while dark spots are observed on the antennas on the contrary. In the inverted-anomalous mode we observe record signal values of $5 \cdot 10^4$ photons per second. We attribute the anomalous effect to the realization of a conditionality for resonant tunneling of electrons with excitation of optical states.

Keywords: golden nanoantenna, femtosecond laser printing, nanoscale on-chip light sourse, luminescence from tunnel junction, inelastic tunneling of electrons, resonant electron tunneling, scanning tunneling microscope

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Усиление люминесценции при неупругом туннелировании электронов за счет изменения геометрии туннельного контакта

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Аннотация. Экспериментально исследовано световое излучение, возникающее в результате неупругого туннелирования электронов в переходе с полусферическими золотыми наноантеннами (d = 400 нм, h = 300 нм), созданными методом прямой фс-лазерной абляции. Были выделены два характерных режима люминесценции: стандартный при 2,5 В – повышенная плотность локализованных оптических состояний (ЛПОС) (оптический сигнал до ~$7 \cdot 10^3$ фотонов в секунду) наблюдалась в области наноантенн и аномальный при 2,7 В, при этом ЛПОС повышена по всей поверхности золота между наноантеннами (пиковые значения до ~$5 \cdot 10^4$ фотонов в секунду), а на антеннах, наоборот, наблюдались «темные» пятна. Резкое увеличение интенсивности излучение ассоциируется с проявлением резонансного туннелирования через оптические состояния.

Ключевые слова: наноантенны, фемтосекундная лазерная печать, наноразмерные источники света на кристалле, люминесценция из туннельного перехода, неупругое туннелирование электронов, резонансное туннелирование электронов, сканирующий туннельный микроскоп

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Introduction

One of the main challenges in the design and successful realization of optoelectronic integrated chips is the development of a nanoscale on-chip photon source whose emission can be excited and controlled by electrical signals and which is compatible with current semiconductor technologies [1].

A promising way for the implementation of such light source is the use of tunneling nanocontact between two metal surfaces (metal-insulator-metal (MIM)) [2]. When electrons tunnel through a MIM contact, light is emitted by inelastic tunneling (LEIT). However, the probability of LEIT (external quantum efficiency (EQE)) process is extremely low, approximately ~10^{-6}–10^{-7}, limiting the prospects for commercial realization of simple planar MIM photon sources [3–4].

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Our previous work [5] demonstrated that due to modifying the surface by gold nanoantennas formed using femtosecond laser printing technique, the EQE increased by an order of magnitude compared to the flat surface. These nanoantennas acted as plasmon resonators, concentrating the electromagnetic field and increasing the local density optical states, thereby increasing the probability of photon emission [6–7].

The main objective of this study is to further enhance the EQE in a MIM structure with gold nanoantennas by varying both the geometrical and energetic parameters of the tunnel gap. By fine-tuning these parameters, we aim to maximise the interaction between the tunnelling electrons and the optical modes of the nanoantennas, thereby improving the photon emission efficiency.

Materials and Methods

The initial sample is a gold film with a thickness of 50 nm deposited on an optically smooth glass substrate by electron beam evaporation. Rows of nanoantennas with a period of 1 to 1.5 μm and a diameter of 500 nm were printed on gold film by femtosecond laser printing technique. SEM image of a cross section of the structure is shown in Fig. 1, *a*. [2]. The nanocontact with the sample was formed using a tungsten chemical etched probe in an ultrahigh vacuum tunneling microscope (Omicron UHV VT AFM/STM). Spatial STM-L maps were recorded using an external data acquisition system based on a single-photon counter with a spectral range of 400–900 nm [3]. Features of the optical assembly for luminescence detection and low total luminescence intensity from the tunnelling contact do not allow obtaining emission spectra for particular wavelengths in this experiment. The optimum tunnelling contact parameters were searched for at a fixed bias voltage of 2.7 V (one of the characteristic luminescence peaks of the structures on the tunnelling VAC) and tunnelling currents in the range of 100-5000 pA by multiple registration of STM-L images. The dependence of STM-L optical signal intensity on bias voltage were taken for voltage variation in the range of –4 to 4 V at a fixed tunnelling current of 1000 pA, the obtained curve is shown in (Fig. 1, *d*). Fig. 1 shows two sets of typical STM and STM-L surface images obtained during the study of the optimal parameters of the tunnel contact: the standard luminescence mode (Fig. 1, *b*, *c*) and the inverted mode (Fig. 1, *e*, *f*).

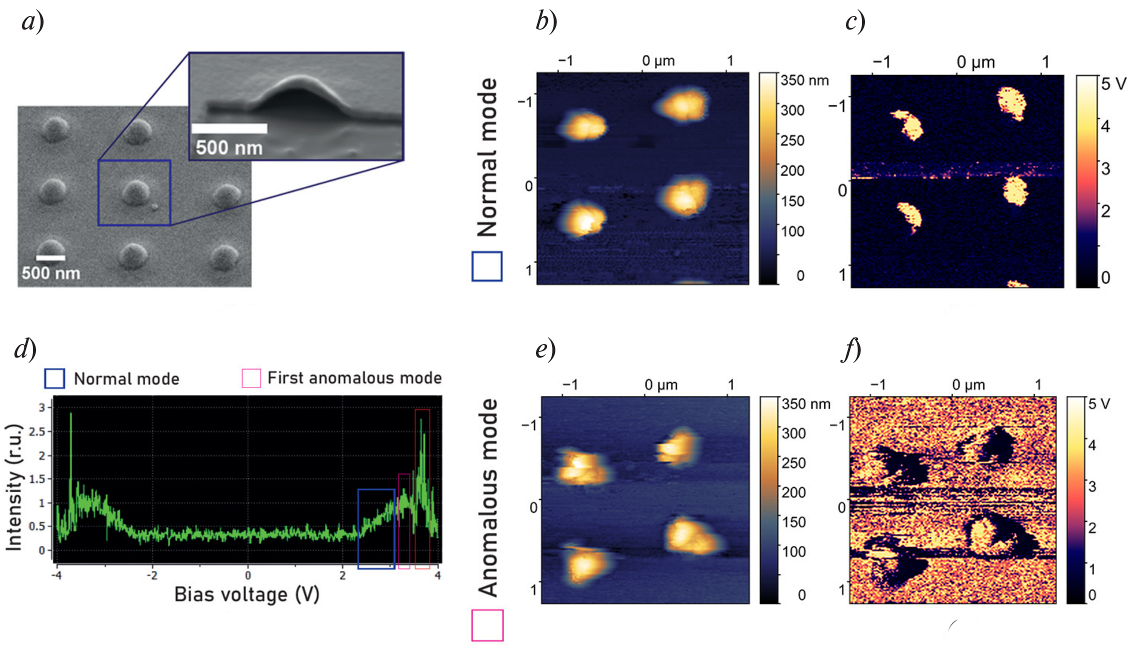

Fig. 1. Synchronized recording of STM topography and luminescence (STM-L): SEM image of cross section of the nanoantenna (*a*); dependence of luminescence intensity on bias voltage at fixed tunnel current of 1000pA (*d*); STM topography of nanoantennas (*b*, *e*); STM-L map of nanoantennas (*c*, *f*)

Results and Discussion

In the whole investigated range of tunneling contact parameters, two modes can be distinguished. First standard luminescence mode, consists in a smooth increase of the signal on the STM-L map with increasing tunneling current up to ~2.25 nA at 2.7V. An increase in tunneling current means that the probe is approaching the surface. This range of bias voltages is highlighted on the intensity curve by a blue frame in Fig. 1, *d*. Further increase of the current at 2.7 V leads to a sharp change of the pattern to an anomalous one. In the anomalous regime, the luminescence (LDOS) increases dramatically over the entire surface area in the STM-L map more than 5 times in amplitude from $7 \cdot 10^3$ photons per second to peak values of $5 \cdot 10^4$ photons per second. At the same time, in contrast, "spots" with reduced LDOS are observed over the nanoantenna region. Further increasing the bias voltage above 3.5V, a smooth decline in the optical signal intensity is observed due to a change in the nature of charge carrier transport from tunnelling to over-barrier transport ("breakdown" of the tunneling barrier). When the current is reduced to 2 nA at 2.7 V or below 2.7 V (at any current), the transition to the normal luminescence mode occurs.

Conclusion

The fact of such a dramatic change in the luminescence pattern and increase in the luminescence intensity, respectively, EQE, may indicate the manifestation of resonant tunneling of electrons with excitation of optical states. The processes of electron tunnelling occurring in the inverted (anomalous) luminescence regime and spectral studies of the emission features require further deeper investigations.

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