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Investigation of ion transport in solid-state nanopores upon optical radiation

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Abstract. We developed a technique for the fabrication of single nanopores with gold bow-tie nanoantennas in a free-standing SiN membrane of arbitrary thickness. Single pores with a diameter of 30–50 nm and a length of 300 nm were fabricated. Studies of ion transport in solid-state nanopores upon optical radiation showed that the enhancement of an electromagnetic field by plasmon structures near nanopores leads to the increase in nanochannel conductivity.

Keywords: ion transport, nanopore, nanochannel, SiN membrane, plasmon structure, gold nanoantennas

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Влияние оптического излучения на ионный транспорт в твердотельных нанопорах, содержащих плазмонные структуры

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Аннотация. Разработана методика изготовления одиночных нанопор с золотыми наноантеннами в тонкой мембране SiN. Получены одиночные поры диаметром 30–50 нм и длиной 300 нм. Исследования переноса ионов в твердотельных нанопорах под действием оптического излучения показали, что усиление электромагнитного поля плазмонными структурами вблизи нанопор приводит к увеличению проводимости наноканалов.

Ключевые слова: ионный транспорт, нанопора, наноканал, SiN мембрана, плазмонная структура, золотые наноантенны

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Introduction

Experimental and theoretical research of transport in micro- and nanochannels not only has fundamental interest, but also carries great significance for applications in various fields of science and technology. These include the separation of solution, purification of substances [1] and development of biochemical sensors [2]. Nowadays, the study of molecular and ion transport in biomimetic microfluidic systems containing nanopores and nanochannels is of increased interest [3–5].

Microfluidic devices with integrated micro- and nanoscale structures can be used to develop devices for the analysis of individual molecules, pre-concentration/separation of samples and single-molecular DNA/RNA sequencing [6]. Nanofluidics-based approach has a number of unique advantages, such as an ultra-high surface-to-volume ratio and scales comparable to a range of different surface/interfacial forces and sizes of biomolecules (DNA, RNA, proteins) [7]. This allows us to explore new transport phenomena that occur only at the nanoscale.

Earlier, our research group developed a method for the formation of microfluidic chips containing a nanopore array [8].

The aim of the current study is to investigate ion transport in solid-state nanopores upon optical radiation. The methods of electron and ion lithography make it possible to create nanopores with a given geometry and specified conductive properties. Such structures are an optimal model system for the study of ion transport.

Materials and Methods

To obtain samples the technique of nanopore formation in a suspended semiconductor membrane was used. This method can be divided into two stages. At the first stage, it is necessary to form a free-standing semiconductor membrane in a silicon substrate. For this, SiNx layer with a thickness of ~300 nm was deposited onto the double-sided polished Si (100) plate by low-pressure chemical vapor deposition (LPCVD). The thickness of this layer determines the length of the future pores. Then, using photolithography, windows were formed in the SiNx layer for further anisotropic etching of Si substrate. As a result, a free-standing silicon-based SiNx membrane was obtained. The membrane had a square shape with a side of 65 microns.

To study the effect of optical radiation on the transport properties of nanopores (nanochannels), a plasmonic bow-tie nanoantenna made of gold were formed (Fig. 1, *a*). The bow-tie structure consists of two triangular platforms pointed at each other by the vertices of triangles. At the second stage, nanopores were formed in the fabricated SiNx membrane within bow-tie antennas by focused ion beam (FIB) milling. The diameter of the synthesized pores was approximately 30 nm (see scanning electron microscopy (SEM) image in Fig. 1, *a*).

Results and Discussion

When the plasmon structure is exposed by optical radiation at a resonant wavelength, the energy of an optical beam is localized within antennas via near field component of electromagnetic field. Thus, by nanopore located between gold plasmon structures, it is possible to study the effect of optical radiation on the ion transport. Figure 1, *b* shows two spectra: the yellow one refers to gold plasmon structure located on the membrane without pore, while blue one refers to plasmon structures with the drilled nanopore. Thus, the presence of pore does not affect the spectrum. The peak of the scattering spectra is located in the wavelength range of 500–700 nm.

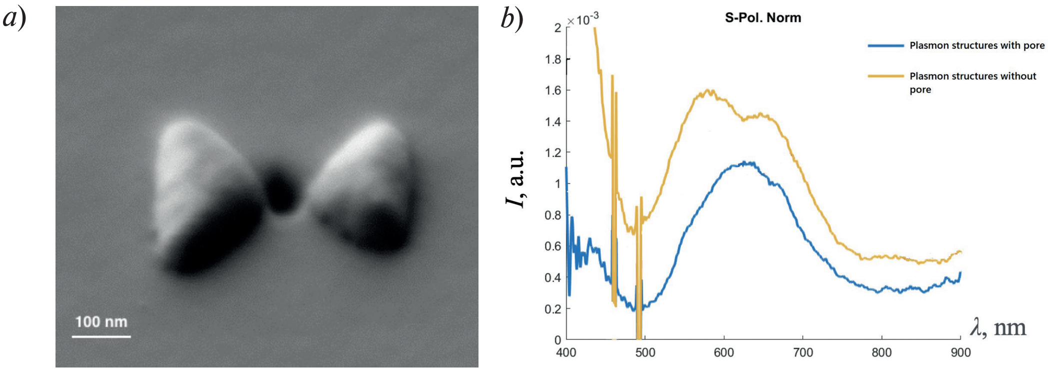

Fig. 1. SEM image of nanopore (nanochannel) modified with plasmon bow-tie nanoantenna on the surface of SiNx membrane (*a*), dark-field scattering spectra of plasmon bow-tie nanoantennas with and without nanopores on the surface of SiNx membrane (*b*)

A measuring cell with optical access (Fig. 2, a) was developed for the further experiments. The cell consists of two electrolyte reservoirs separated by an impermeable SiNx membrane with a single nanopore. An optical window was made at the bottom of the lower half of the cell to place on the microscope slide. Thus, laser beam can be focused in the center between the plasmon antennas using a 50x lens. A 532 nm laser with a power of 6.5 mW was used for the measurements.

To study the effect of optical radiation on the ion transport through the pore, the cell reservoirs were filled with a KCl solution with a concentration of 10 mM and the ion conductivity of the nanopore was studied by measuring the current-voltage (I-V) characteristics (Fig.2, *b*) without laser radiation and with the laser turned on. One can see that the laser irradiation enhances the channel conductivity. This can be explained by the enhancing of local temperature within nanopore, which provides the increase of ion transport.

Silver-chloride electrodes were used for the measuring. It is worth mentioning that the surface of the silver electrodes was refreshed by chlorinating the silver wires in 1M KCl solution before each experiment.

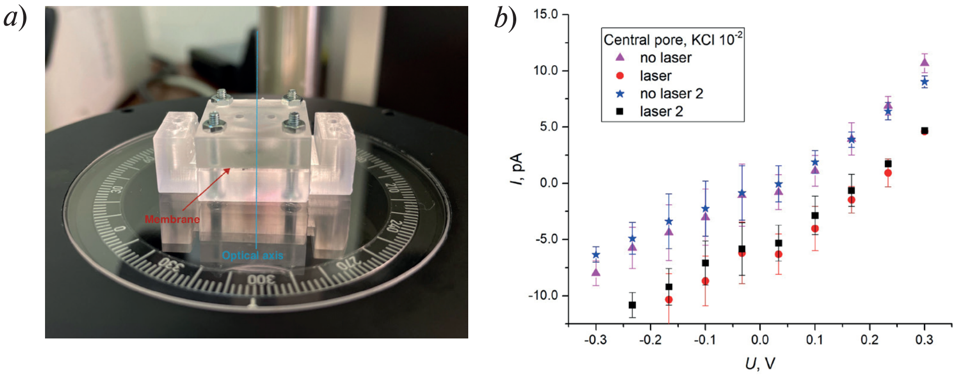

Fig. 1. SEM image of nanopore (nanochannel) modified with plasmon bow-tie nanoantenna on the surface of SiNx membrane (*a*), dark-field scattering spectra of plasmon bow-tie nanoantennas with and without nanopores on the surface of SiNx membrane (*b*)

Conclusion

The conductivity values for two measurement modes without and with laser radiation are 3 nS and 3.5 nS, respectively. We can conclude that the enhancement of an electromagnetic field by plasmon structures near nanopores leads to the increase in conductivity. It is worth mentioning that the conductivity values restore to the initial value while switching the measurement mode.

Such effect can be explained by the local heating of the solution near the pore and a temperature gradient, which, causing Brownian motion of ions, affecting the conductivity. The proposed technology makes it possible to create pores of a controlled size in a free-standing SiN membrane. The manufactured cell with optical access allows us to study ion transport upon optical radiation. However, during the experiments, we encountered difficulties in filling the cell and pores with a solution. To overcome these obstacles surface treatment for hydrophilization was carried out using oxygen plasma.

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