NanoPen: Light-actuated Patterning of Nanoparticles

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Abstract: We introduce NanoPen, a novel technique for flexible, real-time reconfigurable, and large-scale light-actuated patterning of single or multiple nanoparticles such as metallic spherical nanoparticles, semiconducting and metallic nanowires, and carbon nanotubes.

1. Introduction
The ability to pattern nanostructures has important applications in medical diagnosis and chemical sensing. This is typically achieved by creating micro- and nano-arrays of targeted materials or enhancing the detected signal through methods such as surface-enhanced Raman spectroscopy (SERS). In addition, nanoelectronic and optoelectronic device fabrication and nanostructure synthesis benefit from the ability to pattern nanostructures over large areas. Several techniques such as dip-pen nanolithography [1], nanofabrication [2], contact printing [3], and self-assembly [4] have addressed the challenge of patterning nanostructures. However, these techniques lack the capability to create real-time reconfigurable patterns without the use of complicated instrumentatio or processing steps.

In this paper, we introduce NanoPen, a novel technique for light-actuated patterning of nanoparticles such as metallic nanoparticles, nanowires, and carbon nanotubes. Since NanoPen is an optical patterning technique, the shape of the patterned structures can be controlled and manipulated over large areas, in real-time, by reconfiguring the projected light patterns. In addition, the optoelectronic forces used in immobilizing the particles require low optical intensities at the level of commercial light projectors. NanoPen has single nanoparticle patterning resolution and can tune the linewidth and density of immobilized structures by adjusting the exposure time, light pattern scanning speed, frequency, and light pattern intensity and spot size.

2. NanoPen Device Structure, Mechanism, and Characterization
The NanoPen technique is demonstrated using the optoelectroni tweezers (OET) [5] device structure shown in Figure 1a. The NanoPen mechanism consists of two distinct forces: a collection force which is responsible for collecting the particles in the light spot and an immobilization force which strongly attracts the particles and immobilizes them on the OET surface. The collection force is a combination of the dielectrophoresis (DEP) force, which traps the particles in a short range by polarizing the objects and attracting them to the highest field intensity gradient areas, and a flow-based mechanism called light-actuated AC electroosmosis (LACE) [6], which collects the particles over a longer range, concentrating them in a flow dead zone in the center of the light pattern. The immobilization force which is responsible for attracting the particles to the surface is mainly dominated by the DEP force but is also affected by electrophoresis forces due to the particles surface charges.

Figure 1b shows NanoPen immobilization and patterning of a mixture of 60 and 90 nm gold nanoparticles dispersed in a 4 mS/m solution of KCl and DI water. In the beginning, there was no voltage applied to the device and the nanoparticles underwent Brownian motion. Once the voltage was applied (20Vpp at 10 kHz), the nanoparticles were collected in the center of the light spot and were immobilized on the OET surface. The liquid solution could then be removed without damaging the patterned structure. A linewidth of ~3 μm was achieved for a scanning speed of ~5 μm/s. Figure 1c shows trapping, transport, and immobilization of a single 100 nm spherical gold nanoparticle using NanoPen.

3. Large-scale Nanoparticle Patterning and Applications
In the NanoPen technique, the shape of patterned structures can be tuned in real-time by reconfiguring the projected light patterns. Figure 2a shows patterning of 90 nm gold nanoparticles in the form of the “NIH” logo using a light projector setup. Figure 2b shows patterning of 60 nm gold nanoparticles in the form “CAL” characters using a custom digital micromirror display (DMD) setup. Figure 2c shows patterning of 90 nm gold nanoparticles into a 5×6 array over ~200×150 μm² area. One of the applications of NanoPen patterned metallic nanoparticle structures is in the area of surface-enhanced Raman spectroscopy (SERS) sensing. To demonstrate this capability, we dried a solution of Rhodamine 6G (R6G) dye on the surface of the patterned structure in Figure 1b. The Raman signal from R6G molecules was detected using a 3 mW/532 nm laser excitation. Figure 3a inset shows a 2D scan of the detected Raman intensity at 1570 cm⁻¹ of the patterned structure in Figure 1b. The Raman signal from R6G molecules was detected using a 3 mW/532 nm laser excitation. Figure 3b shows the detected Raman signal with all major R6G Raman peaks visible.

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Fig 1: (a) Optoelectronic tweezers (OET) device structure for trapping and immobilization of nanoparticles. In this structure, light patterns interact with a photoconductive layer (hydrogenated amorphous silicon) deposited on an indium-tin-oxide (ITO) transparent electrode to form virtual electrodes. The field gradients formed in the vicinity of the optically defined virtual electrodes interact with the nanoparticles, trapping them in the potential well. In the NanoPen mechanism, the nanoparticles are collected using the DEP force and the LACE flow and are strongly attracted to the surface by the immobilization force (F_imm) consisting of DEP and electrophoresis forces. (b) Real-time patterning of gold nanoparticles (mixture of 60 nm and 90 nm in diameter) using a manually controlled laser spot. (c) Trapping, transport, and immobilization of a single 100 nm gold nanoparticle using NanoPen.

Fig 2: Large area patterning of nanoparticles using NanoPen. (a) Patterning of 90 nm gold nanoparticles in the form of the “NIH” logo using a commercial light projector (1-10 W/cm² light intensity). (b) Patterning of 60 nm gold nanoparticles in the form of “CAL” characters using a custom digital micromirror device (DMD) setup. (c) Formation of a 5×6 array of 90 gold nanoparticles over 200×150 μm² area.

Fig 3: Measurement of Rhodamine 6G (R6G) spectrum using the immobilized gold nanoparticle (mixture of 60 and 90 nm sizes) pattern shown in Figure1b. (a) 2D scan of R6G Raman intensity measurement for the indicated area at 1570 cm⁻¹. The SERS enhancement is evident in the gold nanoparticle “hot spots” areas. (b) Detection of R6G Raman signal using a 532 nm/3 mW laser source with all the major R6G Raman peaks visible.