



Research paper

Single positively charged particle trapping in nanofluidic systems

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ABSTRACT

High-throughput and contact-free trapping of single nano-objects in an aqueous solution is of substantial interest for fundamental and applied research. One of the several trapping methods is geometry-induced electrostatic (GIE) trapping that allows for passive spatial confinement of single nanoparticles in nanofluidic devices. In aqueous environments ($\text{pH} > 2$) glass and silicon dioxide surfaces acquire a net negative surface charge density due to the self-dissociation of terminal silanol groups. Thus, with native glass/silicon-based GIE-trapping devices only negatively charged nano-objects can be trapped, limiting the applications of this method. In this work, we have performed surface modifications of glass-based GIE-trapping nanofluidic devices to enable the trapping of positively charged nanoparticles. For surface functionalization of the devices, a layer transfer of poly-(ethyleneimine) electrolytes were used which provides a net positive surface charge density. We demonstrate the successful confinement of positively charged 60 nm gold nanoparticles inside the functionalized devices, and present a comparison study between trapping of negatively- and positively charged particles in native and functionalized devices, respectively.

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1. Introduction

Contact-free trapping and handling of single nano-objects in solution is of significant interest for potential applications in chemistry, biophysics, biotechnology, and clinical medicine [1]. A contact-free trapped nano-object can be studied in a more controlled manner and for longer time intervals to provide information on local dynamics and on chemical or biological properties of the object of interest. Several active trapping methods, such as optical [2,3], magnetic [4,5], and acoustic tweezers [6,7], have demonstrated successful trapping of single objects ranging from ~ 10 nm to ~ 100 μm in size [3,8]. In these methods, the restoring force is proportional to their respective field gradient, to the volume of the trapped object, and other object properties such as polarizability (α) for optical tweezers, magnetic susceptibility for magnetic tweezers, and compressibility and density for acoustic tweezers [3,8,9]. Reduction in the size of the trapped nano-object attenuates polarizability, susceptibility and other volume dependent particle properties, which results in the requirement of very high field gradients. Thus, these methods are not very effective for smaller objects (< 100 nm). With relatively complex setups, optical tweezers allow for the trapping of nano-objects down to ~ 10 nm [2,3,10–12] and a spatial resolution of

a few Angstroms [13,14]. An alternative to these methods is geometry-induced electrostatic (GIE) trapping, which allows for contact-free confinement of nano-objects down to tens of nanometers [15,16]. GIE-trapping is a passive confinement method in aqueous environments that is achieved through spatially modulated electrostatic potential wells by tailoring the surface of a nanofluidic device. The stiffness and stability of the electrostatic trapping are dependent on the charge of the confined object, the geometry of the system and the concentration of the ionic buffer solution. Thus, it allows for the trapping of various particles ranging from single gold nanoparticles (Au NPs) to lipid vesicles independent of their mass [16]. Significant investigations were performed to achieve the immobilization of various objects in different geometries and orientations using GIE-trapping [17,18].

GIE-trapping nanofluidic devices are mainly based on SiO_2 or glass substrate [15,16]. The presence of silanol groups on selected substrates generates a negative surface charge density when the substrate is in contact with an aqueous solution ($\text{pH} > 2$) due to the deprotonation of terminal silanol groups [19,20]. The resulting negatively charged surface causes the redistribution of coions and counterions in the aqueous solution, which leads to the formation of an electric double layer (EDL) as shown in Fig. 1(a). In the presence of an EDL, the electrostatic potential decays linearly in the Stern layer, and exponentially in the double layer [21], where the electrostatic potential distribution beyond the Stern layer can be expressed by the Poisson-Boltzmann (PB) equation [16].

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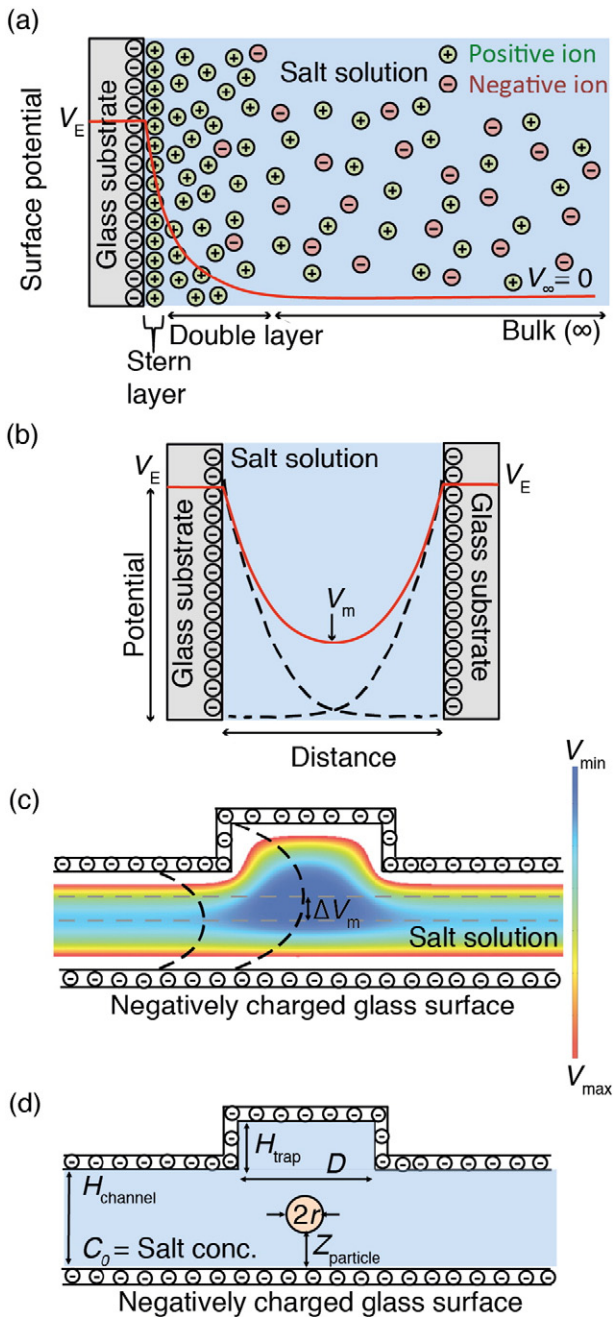


Fig. 1. (a) Stern representation of an electrostatic potential distribution in an aqueous medium due to the self-dissociation of terminal silanol groups of the glass substrate. (b) Generation of a potential minimum in between two charged surfaces in an aqueous medium, from the overlap of two exponentially decaying electrostatic potentials. (c) Shift in the position of the electrostatic potential minimum, and the formation of a local potential well inside a nanopocket in the tailored topographic surface of the GIE-trapping device. (d) Parameters of a GIE-trapping device that directly influence the trapping stiffness of the particle inside the nanopocket.

When two charged surfaces are brought close to each other in a buffer solution, due to an overlap of the electrostatic potentials from both the surfaces, an electrostatic potential distribution is formed, where the minimum of the potential distribution lies in the middle of the two surfaces (Fig. 1(b)). The presence of a potential minimum forces a negatively charged particle to acquire the lowest potential energy position at the mid-plane of the two surfaces. Tailoring the topography of one side of the fluidic device brings a shift in the location of potential minimum away from the mid-plane which creates a local potential

well and thus imposes a confinement of the particle towards the pocket as shown in Fig. 1(c). The difference of the potential energy inside the pocket and at the mid-plane of the two surfaces away from the pocket structure should be sufficiently higher than the thermal energy of the particle for a successful GIE-trapping. The potential distribution in the presence of nanopockets is shown in Fig. 1(c), where the shift in the potential minimum can be seen clearly. The shape of the potential well inside the pocket can be tuned by geometric parameters of a GIE trapping device such as the channel height, trap height, and trap diameter, or by the salt concentration of the buffer solution inside the device as highlighted in Fig. 1(d). A deeper electrostatic well gives high residence times whereas a deeper and steeper potential well allows for a stiffer trapping for the confined particles [16]. To track the motion of a trapped object in the pockets, interference scattering detection (iSCAT) technique [22,23] was employed in the present study. In several excellent studies, iSCAT has been shown to be a powerful technique for the observation of nano-objects such as Au NPs, viruses [24], polymer beads, lipid vesicles [16,18], and motor proteins [25]. It relies on the coherent detection of the interference signal between the reflected light from the fixed geometries and elastically scattered light from the particles. Thus, iSCAT allows for a simultaneous imaging of both the particle and the nano-structured surface.

In contrast to other trapping methods, GIE-trapping devices have been used exclusively for negatively charged objects to date. To employ GIE-trapping devices for positively charged particles, it entails modification of the device surface. Glass-based GIE-trapping devices in their native state allow only the trapping of negatively charged objects. In order to trap positively charged objects, an effective and stable functionalization of the inner surface of the device with a layer of a net positive surface charge density is needed.

In this work, we demonstrate the reliable contact-free trapping of positively charged Au NPs of 60 nm diameter in glass-based nanofluidic GIE-trapping devices. It is achieved by the functionalization of the device surface with positively charged polyelectrolytes to acquire a net positive surface charge density at the inner surface. We characterize the device performance by analyzing the particle lateral movements and compare it to negatively charged Au NPs trapped in the non-functionalized glass-based devices. This work will further expand the possibilities for trapping and handling of nanoparticles and biomacromolecules that have a net positive surface charge.

2. Material and methods

2.1. Fabrication of glass-glass-based GIE trapping device

GIE trapping nanofluidic devices can be achieved from both silicon and glass substrates. However, in silicon-based devices, the highly reflective Si-SiO₂ interface brings in several detection challenges, whereas glass-based devices provide a high signal-to-noise ratio in iSCAT detection technique [15]. Therefore, in this work, we exploit glass-based GIE trapping devices that are fabricated with conventional lithography processes on a 500- μ m-thick borosilicate glass wafer (Borofloat® 33, Plan Optik). A final device consists of two micro- and several nanofluidic channels with embedded nanopockets. The two microfluidic channels are connected by the nanofluidic GIE trapping channels and thus allow in- and outflow of buffer and sample solutions as shown in Fig. 2.

The complete fabrication process was conducted in several steps. To start with, a wafer was cleaned with acetone in a sonication bath for 10 min and then with freshly prepared piranha solution (H₂SO₄ (%):H₂O₂ (%) = 2:1) for 20 min, which was followed by deionized (DI) water cleaning and drying under a nitrogen gas stream (N₂).

To pattern the microfluidic channels, a 5-nm-thick chromium (Cr) layer (adhesion layer), and a 150-nm-thick Au layer were evaporated (Univex 450, Cr rate: 1 Å/s, Au rate: 1 Å/s) on both sides of the wafer.

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