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Research paper

Nanofluidic lab-on-a-chip trapping devices for screening electrostatics in concentration gradients

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ABSTRACT

Geometry-induced electrostatic (GIE) trapping is a novel contact-free method of stably confining charged nano-objects in solution. This method has proven to be very effective in trapping sub-100 nm objects and is based only on the electrostatic repulsion between the charged object and the device surfaces, without requiring an external control or power. We report on fabricating a GIE trapping device integrated into a microfluidic system and demonstrate its performance in screening the behaviour of individually trapped nano-objects along a NaCl salt concentration gradient. We use 60 nm gold particles as probes to analyze the trapping stiffness and residence time of the particles along the salt gradient. We show that in our devices a critical concentration for the reliable trapping of the particles in the order of seconds is reached at an ionic concentration of 0.3 mM. By analyzing the trap stiffness and residence times, we determine a smooth gradient of the salt concentration, as expected from Fick's first law. Furthermore, we find that the instability of the colloidal dispersion is reached at 0.8 mM NaCl.

1. Introduction

Stable contact-free trapping and detection of single nano-objects in solutions provide the ultimate sensitivity in characterizing analytes at nanometer dimensions. This methodology offers explicit information on local dynamics, reactions, structural information, and net charges. Extensive developments in active trapping methods, such as optical [1–3], plasmonic [4–6], magnetic [7,8] or acoustic [9,10] tweezers, rely on the induced field gradient and have been successfully demonstrated for stable confinement of single objects. However, stable trapping of nano-objects smaller than 100 nm remains challenging for such methods as the trapping force, for instance in optical tweezers, relies on the polarizablility, α, of the trapped objects, which scales with the third power of the object size. In addition, α vanishes when the material properties of the object and its surrounding media are similar. As a result, for trapping ever smaller objects using optical tweezers, large field powers in the order of hundreds of mW are needed [11], which might lead to photodamage in specimens [12].

Geometry-induced electrostatic (GIE) trapping is a field-free method that allows reliable confinement of nanometer-size objects for time durations from seconds to hours [13]. This method comes with the key benefit that the trapping force depends only on the net charge of the object rather than on its size and mass. In conventional GIE trapping devices, single negatively-charged nano-objects are trapped by electrostatic repulsion from negatively-charged SiO_2 walls in nanofluidic channels. The surface topology of the channels is tailored by nanometer-sized indentations, which results in the formation of local energy potential wells, as shown in Fig. 1. This system has the flexibility that various trapping geometries such as circular pockets or rectangular slits or grids can be realized using, for example, e-beam lithography. The depth of the potential wells, and thereby the trapping strength and time, can be adjusted by altering the size of the indentations, the nanofluidic channel height, the surface charge density, and the ionic concentration of the buffer solution. GIE trapping has evolved over the years to be used for stable trapping of single gold nanoparticles (Au NPs) [13–15], polymer beads, lipid vesicles [13], as well as for angular dependent trapping of silver nanorods [16], which can be used for digital information storage applications [17]. While lab-on-chip approaches made from silicon or glass substrates have fixed nanofluidic channel heights and trap geometries, active approaches such as scanning-aperture trapping [18] or piezo-controlled SiO_2 slices [19] are capable of altering the trap

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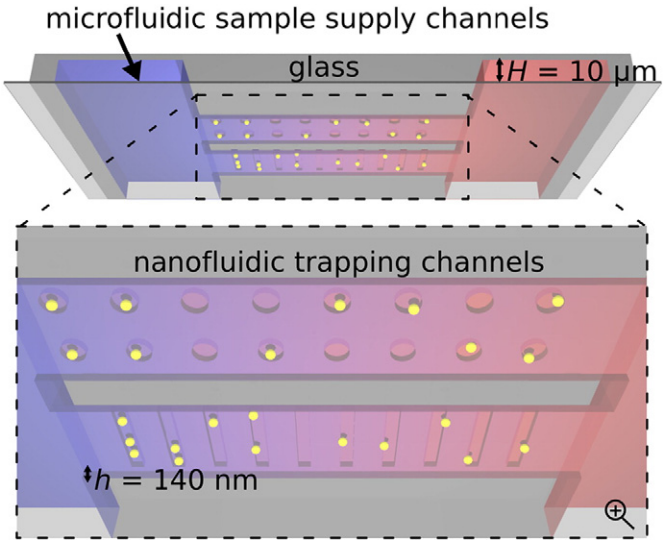

Fig. 1. Schematic of a geometry-induced electrostatic (GIE) trapping device integrated into a microfluidic system. Nano-objects are trapped within the pockets and slits of the nanofluidic channels (middle) by electrostatic repulsion. Large microfluidic supply channels provide the GIE trapping area with reactants or buffer solutions (left and right).

potential depth during the experiment. Nevertheless, the ease-of-use of the chip-based devices makes integration into more complex microfluidic systems straightforward and would provide a range of new applications for electrostatic trapping, such as high-throughput screening of nano-objects, *in situ* mixing and sorting of nano-objects, or the feasibility to precisely control the fluidic conditions.

In this work, we demonstrate the successful integration of GIE trapping devices into a microfluidic system and utilize it for determining very small sample and reagent quantities, precise control of reactant concentrations, and short analysis times (see Fig. 1). In this system, a precise and controlled steady-state concentration gradient can be formed along the trapped nano-objects e.g. of reactants, pH or salt, allowing the analysis and screening of contact-free trapped nano-objects in different environments in a single experiment using one device. We use Au NPs of 60 nm diameter as probes along a NaCl gradient, and thereby find critical concentrations on trapping strength and particle aggregation stability, an important parameter of nanoparticle emulsions. For optical visualization of the particle motions, we used interferometric scattering detection (iSCAT), which relies on the interference between the reflected beam at the interfaces in the device and the light scattered from the particle. Due to the strong optical scattering of the Au NPs [15] and the interferometric nature of the detection, iSCAT allows for high signal-to-noise ratio (SNR) imaging and thus sensitive and fast detection of particle trajectories [13,15,20,21]. For improved SNR imaging [22], the devices were fabricated from glass substrates using top-down nanofabrication methods, i.e. reactive ion etching (RIE) and electron beam (e-beam) lithography.

2. Chip design and working principle

The architecture of the multi-height device is sketched in Fig. 2A. It is made of two main elements: the microfluidic channels, which serve as sample supplies, and the GIE trapping nanofluidic area. Two microfluidic channels are separated by a distance of 890 µm and have a depth of $H = 10$ µm, each connected by an inlet and outlet. The two microfluidic channels are interconnected by several GIE trapping channels with a length of $L = 890$ µm and a height of $h = 50$–200 nm. Within these nanofluidic channels finer nanostructures, i.e. the actual nanotraps are etched. Microfluidic tubings are connected to the inlets and outlets for the delivery of sample, reactant, and buffer solutions.

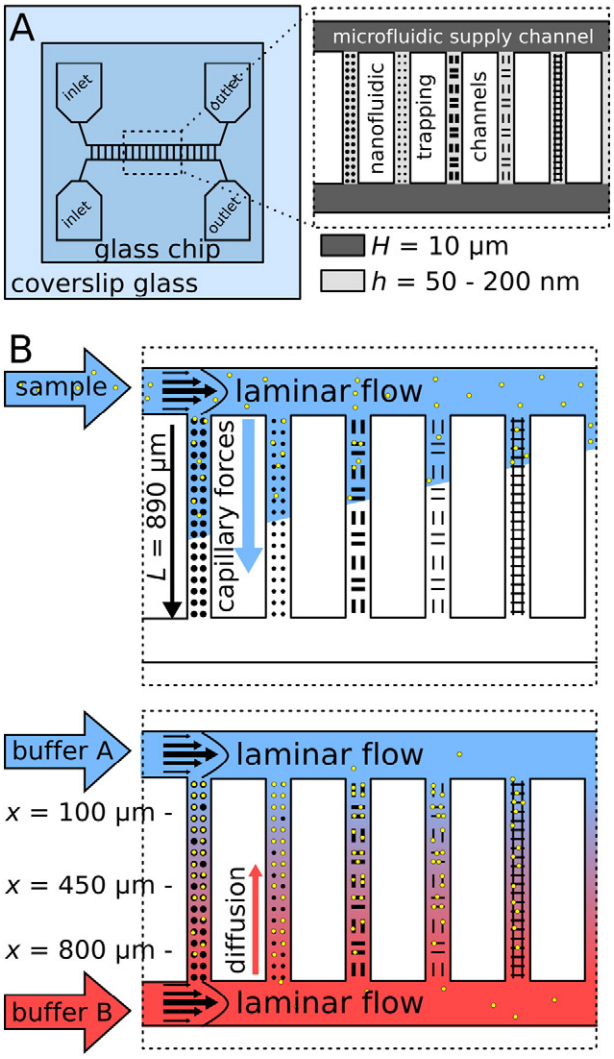

Fig. 2. A) Top view schematic of the microfluidic device design with the integrated nanofluidic GIE trapping area. The microfluidic channels have a depth of 10 µm and are only connected through the nanofluidic channels with a height from 50 to 200 nm. B) Sketch of the experimental process and the principle function of the device. The nano-objects are filled into the GIE trapping area by capillary forces. A linear steady-state gradient in the trapping region (nano-channels) is created by flowing two different solutions through the upper and lower microfluidic supply channels, respectively. The gradient is formed in the nanofluidic channels by diffusion. Thus the analysis of individual nano-objects at different concentrations is achieved within a single device.

The device is filled by injecting the sample solution through the upper microfluidic supply channel as sketched in Fig. 2B. The GIE trapping nanofluidic channels are easily filled with the sample solution containing the NPs by capillary forces. After the particles are trapped in the GIE trapping area, the upper and lower supply channels are flushed with buffer and reactant solutions, respectively. The supply channels are only connected through the nanofluidic channels and are continuously flushed with fresh solutions at equal flow rates to ensure a pressure difference between the supply channels of $\Delta p = 0$. This results in a linear steady-state gradient between the two buffer solutions in the GIE trapping area, caused by diffusion [23]. Based on Fick's first law of diffusion, the net flux of the ions or reactants in solution in the nanofluidic channels, J_x, is written as [24]

$$J_x = -D\,\frac{\partial C}{\partial x} = -D\,\frac{C_2 - C_1}{L}, \tag{1}$$

where D is the diffusion coefficient and $\frac{\partial C}{\partial x}$ is the change of concentration along the nanofluidic channel between the upper and lower solutions