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Glass-based geometry-induced electrostatic trapping devices for improved scattering contrast imaging of nano-objects

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ABSTRACT

Trapping of micro- and nano-objects in solution is of great scientific interest in various fields. One method of trapping and detecting objects smaller than 100 nm is the combination of geometry-induced electrostatic (GIE) trapping devices and interferometric scattering detection (iSCAT). In GIE trapping, charged nano-objects are confined in a nanofluidic system that hosts topographically modified surfaces, resulting in electrostatic potential wells. We observe optical limits of detecting gold nanoparticles smaller than 60 nm because of the high reflection of the strong background signal in current silicon-based GIE trapping chips. The high reflection rapidly leads to overexposure of the camera detector and thus limits the incident laser power. In this work, we introduce new functional geometry-induced electrostatic devices fabricated from glass substrates. Due to the reduced reflection at the water–glass interface compared to the silicon-based devices, higher incident laser power can be used to image the nano-objects resulting in higher contrast as well as signal-to-noise ratios (SNR) of the gold nanoparticles. Using glass-based GIE trapping devices, significant SNR increases are achieved in comparison to that of silicon-based devices. These improvements enable the detection of much smaller nanoparticles and thereby studies on their trapping, as well as further investigation in nanofluidic systems.

1. Introduction

Contact-free trapping of nano-objects is of broad interest for a range of disciplines such as biotechnology, biophysics and material science. Methods such as optical- [1–3], magnetic- [4], and acoustic- [5] tweezers or electro- and dielectrophoresis [6] are successful methods that have been demonstrated. However, these methods come with the limit of the restoring force, $F_{res} \propto \alpha|\nabla E|^2$, where $|\nabla E|$ is the field gradient, and α is the polarizability of the trapped object. α vanishes in the limit of small particles or when the material properties are similar to the surrounding media. Moreover, to get a stable trapping of small nano-objects, large gradients and fields have to be applied. In addition to often demanding setups, large fields might damage the object of interest. A promising candidate for trapping objects smaller than 100 nm in fluid without any externally applied fields is geometry-induced electrostatic (GIE) trapping [7–10]. Contrary to the mentioned trapping methods, F_{res} in electrostatic trapping is proportional to the object charge rather than to the size and mass. Therefore, this method has a great potential in polymer physics [11,12], protein and DNA analysis [13], and as a sample environment for single protein free electron laser (FEL) experiments. Altering surface topology of nanochannels in silicon-based chips was used in a first realization of GIE traps [7]. It was shown that by creating localized three-dimensional potential wells, negatively charged nano-objects can be trapped by pockets or be confined to small grooves due to the suppression of their Brownian motion. Gold nanoparticles (NPs) down to 80 nm [8] and fluorescently labeled vesicles in suspension could be confined in electrostatic traps by falling into local potential wells for a time period of several seconds to hours [7,14]. By analyzing the motion of the trapped gold NPs, the size of single gold NPs could be determined in nanometer precision as well as their net carrying charge [14]. Furthermore, orientation-dependent trapping of silver rods could be achieved using this method [9]. A major advantage of GIE trapping is that the potential depth depends only on the charge but not on the mass or size of the nano-object. The electrostatic potential depth can be adjusted by altering the geometry of the chip or by changing the concentration of the buffer solution. A modification of this method is scanning-aperture trapping [10], a tweezing equivalent of GIE traps. This method has the advantage of altering the trap potential depth by

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approaching or releasing the aperture during the experiment and having the option of *x*-, *y*-position scanning, while it is limited to a single trap and a more complex experimental setup.

In most of these studies, charged gold NPs are the prime approach used for characterizing the performance of the electrostatic traps. In contrast to fluorescence labeled molecules, quantum dots (QD) or silver NPs, gold NPs are not limited by photobleaching over time or oxidization and are not limited in time resolution for tracking particles by optical saturation and photoblinking [15]. Due to the strong plasmon resonance peak of gold NPs in the water, in the wavelength range of $\lambda = 530$–550 nm, they exhibit a high scattering signal. However, the detection of gold NPs becomes increasingly difficult with decreasing particle diameter D, since the scattering intensity scales with D^6 [16]. To overcome this drawback, a new method was introduced using coherent light illumination with an interferometric detection scheme [17–20]. This technique, called interferometric scattering detection (iSCAT), is based on the interference between the scattered light of the particle and a reference beam. Very small objects can be imaged by iSCAT at high speeds and with nanometer precision, since the detected signal of objects scales D^3 [17–19]. Using this method, fixed gold NPs of 5 nm size at a glass–oil interface using supercontinuum white light [17] and at a glass–water interface using confocal scanning iSCAT were detected [18]. Due to its high sensitivity in the phase difference between the reference and scattered beam and the consequential contrast change in axial movements of the particles, iSCAT was used to track 80 and 100 nm gold particles with less than 5 nm localization precision in three dimensions at an acquisition rate of 1 kHz [7,14]. The first concrete application of iSCAT for single biomolecule tracking was shown by Kukura et al. [19]. By simultaneously detecting the fluorescence signal of a quantum-dot labeled virus and its center of mass by iSCAT, it was possible to track the position and orientation of the virus moving along a lipid bilayer [19]. These studies show the tremendous potential of iSCAT for detection with high sensitivity and for fast imaging with high localization precision.

In this work, we introduce an improved nanofluidic GIE trapping device for better detection of small particles. We focus on the optical detection of these small objects and show the detection of gold NPs down to 40 nm. In this regard, we modify the nanofluidic system to suppress the high reflections and thus limited incident laser beams occurring in silicon-based GIE trapping devices using glass-based chips (see Fig. 1).

1.1. Theory

The basic principal of iSCAT for the detection of small objects at the water–glass interface is illustrated in Fig. 2A. The incident beam E_i, focused to the location of the sample, is reflected at the water–glass interface with the reflected beam E_{r1} and scattered from the particle with the scattered field E_s. The scattered field at the detector is described as $E_s = sE_i$ where $s = |s|e^{i\varphi}$ scales with the polarizability α of the particle [17],

$$s(\lambda) = \eta\alpha(\lambda) = \eta\epsilon_m(\lambda)\frac{\pi D^3}{2}\frac{\epsilon_p(\lambda) - \epsilon_m(\lambda)}{\epsilon_p(\lambda) + 2\epsilon_m(\lambda)}, \quad (1)$$

with the complex dielectric constants of the particle $\epsilon_p(\lambda)$ and the medium $\epsilon_m(\lambda)$ and the proportionality constant η. The total field at the detector E_{det} is given by the sum of the scattered and reflected fields, $E_{det} = E_{r1} + E_s$. The corresponding intensity I_{det} can be written as [7,17–20],

$$I_{det} = |E_r + E_s|^2 = |E_i|^2\{r^2 + |s|^2 - 2r|s|\sin\varphi\}, \quad (2)$$

where the first term scales with the reflected field r^2. This term represents the background intensity, which arises in the case of the gold NP on the glass surface, from the reflected beam at the glass–water interface, E_{r1}, depicted in Fig. 2A. The second term, proportional to $|s|^2$, represents the purely scattering signal, which scales with D^6 and thus becomes smaller than the noise of the field reflectivity for very small particles. The last term $2r|s|\sin\varphi$ is the interference of the reflected and the scattered field and scales with D^3. For very small particles, this term dominates the pure scattering signal, since it is multiplied by the reflected field r. A significant advantage of iSCAT compared to, e.g., fluorescence microscopy is that at low intensities the incident beam power can be simply increased since the scattered field of the particle increases with the incident beam. Fluorescence microscopy is here limited by the fluorescence saturation of the dye and thus longer integration times.

The above argument is true for particles sitting on a glass surface (Fig. 2A) due to the low reflectivity of the glass–water interface. Here, the background intensity, E_r, originates from the reflected field at the glass–water interface E_{r1}, that according to Fresnel's law, has a low reflectivity of $R = 0.2\%$.[1] For current

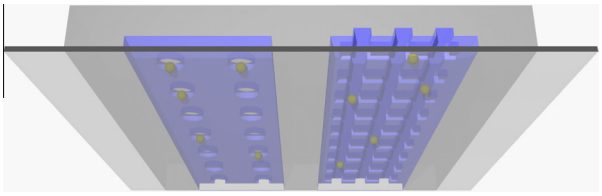

Fig. 1. Schematic of a glass-based geometry-induced electrostatic trapping device. Nano-objects are trapped by the fine structures in the microchannel.

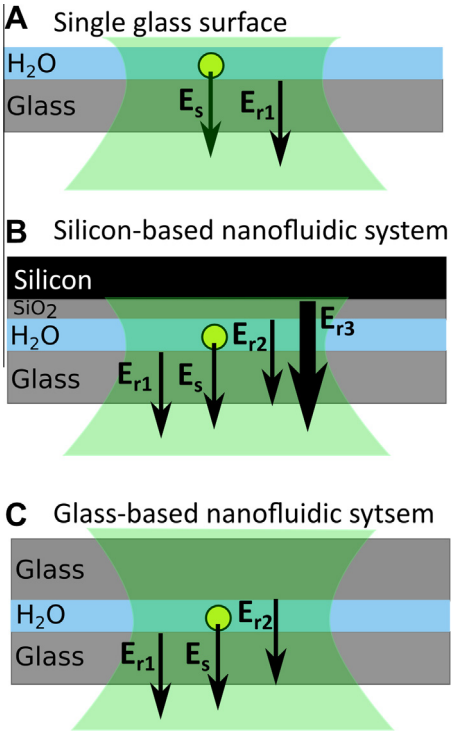

Fig. 2. Schematics of the three nanofluidic devices showing the path of the incident beam, the field scattered from the nano-object and reflected beam at the different interfaces. (A) Nano-object on a single wetted glass surface, (B) current silicon-based nanofluidic systems with a high reflection at the Si–SiO_2 interface, (C) new glass-based nanofluidic systems for GIE trapping with reduced reflection of the incident beam.

[1] Using Fresnel's law for an incident light angle of 90°, $R = \frac{n_2 - n_1}{n_2 + n_1}$, and the refractive indices of $n_{H_2O} = 1.33$, $n_{borofloat} = 1.47$, $n_{SiO_2} = 1.55$ and $n_{Si} = 4.15$.