All-dielectric nanotweezers for trapping and observation of a single quantum dot

ZHE XU¹ AND KENNETH B. CROZIER¹,²,*

¹School of Physics, University of Melbourne, Victoria 3010, Australia
²Department of Electrical and Electronic Engineering, University of Melbourne, Victoria 3010, Australia
*kcrozier@unimelb.edu.au

Abstract: We report the optical trapping of a single streptavidin-coated CdSe/ZnS quantum dot whose overall diameter is around 15–20 nm, in a microfluidic chamber by an all-dielectric (silicon) nanotweezer with negligible local heating. The use of fluorescence microscopy allows us to readily observe trapping events, tracking the fluorescence emission from, and the position of, each individual trapped quantum dot as a function of time. The blinking behavior of the quantum dots is observed during the trapping process, that is, in the near field region of the silicon nanoantenna. We furthermore show that the continuous wave infrared laser employed to trap the quantum dots can also excite photoluminescence from them via two-photon absorption. We present Maxwell stress tensor simulations of optical forces applied to a single quantum dot in the nanoantenna’s vicinity. This work demonstrates that all-dielectric nanotweezers are a promising means to handle quantum dots in solution, enabling them to be localized for observations over extended periods of time.

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1. Introduction

Plasmon-based nano-optical tweezers [1–8] have been shown to offer advantages over conventional optical tweezers [9–11] for the trapping of nanometer-sized objects via the enhanced optical forces that occur when light is confined on a deeply subwavelength scale. The ability to optically trap and manipulate nanomaterials presents many opportunities for nanoscience [12,13]. However, the high level of Joule heating from dissipative losses is recognized as a big challenge faced by plasmonic tweezers [14,15], especially for the trapping and sensing of biological specimens [16]. Loss is also a major issue for many other applications of plasmonics (and not just nanotweezers). This has motivated recent advances in an alternative approach for nano-optics that uses subwavelength high refractive index dielectric nanoelements. These devices are termed all-dielectric nanoantennas [17–23], and generate strong near-field hotspots with much lower absorption losses than plasmonics. In this work, we demonstrate that all-dielectric nanoantennas allow single quantum dots to be trapped optically in a robust fashion.

Colloidal quantum dots (QDs) are nanoscale semiconductor crystals [24] with applications ranging from consumer products such as displays to use as fluorescent markers in biological labeling [25]. They have excellent fluorescence properties [26] that include size-dependent emission, high quantum yield, large linear and nonlinear absorption cross-sections and excellent photostability. At present, there has been much interest in using optical tweezers to trap nanomaterials in solution for a variety of reasons, including to localize these materials to facilitate their characterization via spectroscopy [12,13]. It has been demonstrated that conventional single light-based optical tweezers can be utilized to trap individual QDs and aggregates of QDs [27–33]. This prompts the question of whether QDs can be trapped using nanotweezers supported by surface plasmons and other methods, due to the potential advantages that include tighter spatial confinement, lower optical power requirements and integration with sensing. The trapping of QDs using silicon nitride photonic crystal resonators
has been investigated [34], but this approach is not trivial from an experimental standpoint, requiring tunable laser sources and coupling to waveguides. It has been shown that plasmonic nanotweezers can trap individual QDs and silica-coated QDs [35–37], but it is well-established that the optical properties of QDs can be considerably altered when they are in the vicinity of plasmonic nanostructures [38,39]. This motivates us to demonstrate QDs trapping with a non-plasmonic platform.

In this work, we present direct experiments of optical trapping of a single streptavidin-coated quantum dot by an all-dielectric nanotweezer, that is, silicon (Si) nanoantenna with a Si substrate, via illumination with an infrared wavelength 1064 nm continuous wave (CW) trapping beam. The QD has a CdSe core and ZnS shell, and is further coated by a polymer shell and then streptavidin. According to the manufacturer, the QD has an overall diameter of ~ 15–20 nm. Our simulations indicate that the temperature rise of the nanoantenna due to the trapping laser illumination is negligible and we thus expect no thermal quenching of the QD emission. Employing fluorescence microscopy, we monitor the optical trapping and release of the QD when the trapping laser beam is turned on and off. We furthermore track the trapped QDs versus time, monitoring the fluorescent intensity and centroid position. We observe the fluorescence blinking of single QDs trapped by our silicon nanoantenna. We demonstrate that the illumination from the infrared laser not only results in the optical trapping of a QD, but also excite fluorescence from it via two-photon absorption (TPA). Finally, we quantify the optical trapping forces exerted on the QD using numerical simulations via the Maxwell stress tensor method. This paper illustrates that all-dielectric nanotweezers allow QDs to be trapped optically, without the challenges that can be associated with plasmonic approaches that include local heating and modification to the optical properties of the QDs.

2. Design and simulation

![Fig. 1. (a) Scanning electron micrograph (SEM) picture (45° tilt) of a Si nanoantenna situated on a Si substrate. (b) Temperature rise profile (for steady state) around a Si nanoantenna, \( I_0 = 15 \text{ mW/µm}^2 \). Scale bar: 200 nm. (c) Electric fields profile in the \( yz \)- and \( xz \)-cross section, for 1064 nm plane wave (x-polarization) at normal incidence. White dots represent the origin of coordinate system. Scale bar: 200 nm.](image)

Our all-Si nanoantenna consists of two identical Si nanocylinders separated by a narrow gap (50 nm) and surrounded by a Si ring [Fig. 1(a)]. The two cylinders have diameters and heights of 200 nm. The ring has inner and outer radii of 525 nm and 725 nm, respectively (i.e., is 200 nm wide). The ring has a height of 200 nm. We fabricate the Si nanoantennas via electron beam lithography (EBL) and reactive ion etching methods [23]. We perform finite element method simulations (FEM; COMSOL Multiphysics) of our Si nanoantenna to understand its trapping behavior [23]. In these simulations, the nanoantenna is covered by water \( n_{\text{water}} = 1.33 \) and illuminated at normal incidence by a plane wave (wavelength 1064 nm) with x-polarization from water side (travelling in +z axis). In Fig. 1(b), we plot the steady state temperature rise of the Si nanoantenna for an illumination trapping laser intensity of \( I_0 = 15 \text{ mW/µm}^2 \), which is typical of the levels we employ in our QDs trapping experiments. It can be noticed that the temperature increase is below 0.12 K. This represents a small value, which originates from the fact that Si has a low imaginary permittivity component [40] and a high
thermal conductivity [6]. We thus do not expect to see the quenching or bleaching of the QD emission that can occur with large increases in temperature [19,21]. We next plot the simulated electric field distributions on two cross sections through the nanoantenna as Fig. 1(c). It can be seen that there is a hotspot in the gap between two cylinders, which we attribute to be associated with the coupling between the dipole modes of two Si cylinders [18]. We expect QDs to be trapped here, via the gradient forces accompanying these confined local fields [41,42]. It can be seen that there are also hotspots at the outer extremities of cylinders, but with the smaller electric field enhancement than the hotspot in the gap region between the cylinders.

Figure 2(a) schematically illustrates the structure of the QDs we use in experiments, based on information from the manufacturer (Life Technologies, Q10121MP, Qdot 655 Streptavidin Conjugate) [43]. The QD has a CdSe core and ZnS shell, and is further coated by a polymer layer and streptavidin, with typically 5–10 streptavidin molecules for each QD according to the manufacturer. The QD is ~ 15–20 nm in total diameter. We choose to trap these QDs because they represent a typical choice for biological experiments in which QDs are employed as labels [31,32]. As the QD structure is complex and not known precisely by us, approximations are made when modeling it. Specifically, as depicted in Fig. 2(b), the QD is modeled as consisting of a spherical core (CdSe) with two spherical shells (ZnS and polymer), that is, without streptavidin molecules. The radii of the core and two shells are taken as \( r_1 = 5 \) nm, \( r_2 = 7 \) nm, and \( r_3 = 10 \) nm. We take the refractive index of the outer shell to be 1.6, as this is a typical value for polymers. In Fig. 2(c), we depict the electric field profile on the \( xy \)-plane (\( y = 0 \) cross section) in which the QD is in the gap, centered at \((x_{\text{center}}, y_{\text{center}}, z_{\text{center}}) = (0, 0, -140 \text{ nm})\). For comparison purposes, we provide the calculation for the situation of a single polystyrene nanosphere (NS) with a 20 nm diameter at the same position in Fig. 3. It can be seen that the presence of the QD modifies the fields distribution in the nanoantenna gap more strongly than the NS does. This is because the QD has a larger
refractive index \(n_{\text{CdSe}} = 2.54, n_{\text{ZnS}} = 2.29\) than the NS \(n_{\text{NS}} = 1.6\). As we discuss next, the associated optical forces are also enhanced.

The enhanced and spatially confined field distributions provided by the nanoantenna permit the efficient trapping of single nanoparticles. The general way of computing the optical forces applied to a small object is the Maxwell stress tensor (MST) method \[41,42\]. Here we determine the electric and magnetic fields distributions via FEM simulations and use the MST method to predict the optical forces \[8,23\] exerted on a single QD with the multilayer structure \(r_1 = 5 \text{ nm}, r_2 = 7 \text{ nm}, \text{ and } r_3 = 10 \text{ nm}\). Optical forces are calculated on the polymer surface enclosing the QD. The results are displayed as Fig. 4. In Fig. 4(a), we show the force component \(F_z\) on the nanoparticle versus its center location along the \(z\)-axis. In addition to the QD, we also model a polystyrene NS (with a diameter of 20 nm) and a smaller QD (with \(r_1 = 3 \text{ nm}, r_2 = 4 \text{ nm}, \text{ and } r_3 = 5 \text{ nm}\)). It can be seen that these display similar characteristics, but with the optical force being largest for the QD, followed by the 20 nm NS and the smaller QD. In all cases, the force magnitude is maximum at the nanoantenna edge. It can be seen that when the particle is away from the nanoantenna \((z_{\text{center}} < -200 \text{ nm})\), \(F_z\) is positive, meaning that it is pulled toward the nanoantenna. This continues when the particle enters the nanoantenna region \((-200 \text{ nm} < z_{\text{center}} < -140 \text{ nm})\) until the particle reaches roughly the nanoantenna midpoint, beyond which \((z_{\text{center}} > -140 \text{ nm})\) \(F_z\) is negative. This location \((z_{\text{center}} = -140 \text{ nm})\) thus represents an equilibrium position, that is, the particle will experience a restoring force if it displaced along the \(z\)-axis from this point, for example, by Brownian forces from water molecules. We then plot the horizontal components of optical forces applied to a single QD versus its center location along the \(x\)- [Fig. 4(b)] and \(y\)-axes [Fig. 4(c)]. It can be seen from Fig. 4(b) that \(F_x > 0\) for \(x_{\text{center}} > 0\), and \(F_x < 0\) for \(x_{\text{center}} < 0\). The QD is therefore predicted to have a tendency to be drawn to the sides of the gap. It can be seen that \(F_z\) is positive for the range of \(x\)-axis positions considered, meaning that the QD is drawn to the nanoantenna. This is also the case for the range of \(y\)-axis positions considered in Fig. 4(c). It also can be seen from Fig. 4(c) that the QD feels a restoring force for small displacements along the \(y\)-axis about \((x_{\text{center}}, y_{\text{center}}, z_{\text{center}}) = (0, 0, -140 \text{ nm})\) and \((0, 0, -200 \text{ nm})\). In other words, \(F_y\) is negative for \(y_{\text{center}} > 0\), and positive for \(y_{\text{center}} < 0\).

3. Experimental methods

We next describe our experimental methods and results. The QDs (Life Technologies, Q10121MP) are homogeneously suspended in 1X PBS (phosphate-buffered saline, pH 7.4) solution. Tween 20 (0.05% v/v) has been added as surfactant to avoid aggregation. The concentration of QDs in the experiments is \(\sim 10^8 \text{ mol/L}\). Soon after being made, the suspension is ultrasonicated using a bath sonicator, filtered to remove aggregates, etc. and injected into a microfluidic chamber that is \(\sim 15 \mu\text{m}\) thick. The microfluidic chamber consists of the Si chip containing nanoantennas sealed to a glass coverslip on which a patterned
photoresist layer (~ 15 μm thick) has been formed [23]. Our setup includes an inverted optical microscope (Nikon TE2000), an oil immersion objective (Nikon, 100×, NA = 1.3), a green laser (wavelength 532 nm, for emission excitation), an x-polarized CW infrared laser (for optical trapping, spot diameter ~ 1.2 μm, wavelength 1064 nm), a set of filters, a sensitive electron multiplying camera (EM-CCD, frame rate of 30 fps, exposure time of ~ 30 ms) and a grating spectrometer [23]. This system allows us to observe an individual QD and to track its position as it interacts with the Si nanoantenna. Using this system, we readily observe the optical trapping of a single QD by our Si nanoantenna. Two example experiments are presented as Figs. 5 and 6. These are discussed in the paragraphs below. Illumination intensities of I₀ = 17.3 mW/μm² and I₀ = 16.1 mW/μm² are used for Figs. 5 and 6, respectively. Additional trapping experimental data are provided in Appendix (see Fig. 7). Our experiments confirm that the QDs are optically trapped when the trapping beam is switched on and that they are released from the Si nanoantenna when it is switched off. As shown in Appendix (see Fig. 8), QDs sometimes become stuck to the Si surface. These QDs are readily identified as they are not released from the antenna surface when the trapping beam is turned off. The permanently-adsorbed QDs also appear brighter in the fluorescence microscope images than the optically trapped QDs, making them easy to distinguish.

4. Results and analysis

![Graph](image1)

**Fig. 5.** (a) Fluorescent emission versus time for the trapping of a single QD (see also Visualization 1). (b) Corresponding emission counts histogram of optically trapped QD (from 4–45 s), collected from data of panel (a). (c) Zoom-in of (a) from 35–40 s showing the QD blinking. (d) Four selected EM-CCD images illustrating fluorescence blinking from single trapped QD. Scale bar: 800 nm. (Trapping laser intensity I₀ = 17.3 mW/μm²)
Figure 5 represents one of the key findings of this paper, showing the direct observation of the trapping of a single QD via optical forces in PBS solution using an all-Si nanotweezer/nanoantenna (see Visualization 1). In Fig. 5(a), we depict the fluorescent intensity counts as a function of time, obtained by integrating over a cross section covering 30×30 pixels of each EM-CCD frame that is centered over the Si nanoantenna. It can be seen that there are large fluctuations in the fluorescence signal from the single QD trapped in Fig. 5(a). We attribute this to blinking of the QD [31,33], and note that this was not observed in previous works on the optical trapping of single fluorescent nanoparticles (i.e., polystyrene NSs) [8,23]. We do not observe any quenching or photobleaching of the QD emission [32,37]. This is reasonable because the Si nanoantennas produce ultra-low heat generation [19,23], so quenching or photobleaching associated with the temperature rise should be negligible. It can also be observed that QD fluorescence blinking occurs during the time interval over which the QD is optically trapped (i.e., ~4–47 s and ~49–51 s). Figure 5(b) plots an emission count histogram of the fluorescence blinking data (4–45 s) in Fig. 5(a). Further insight into the blinking phenomenon can be obtained from Fig. 5(c), which represents a zoom-in of the period of 35–40 s [from Fig. 5(a)]. It can be seen that the QD is cycling between emitting “on” and non-emitting “off” states. Four selected EM-CCD frames showing the single trapped QD is given as Fig. 5(d) to further illustrate the blinking process.

We next discuss our results on exciting fluorescence from individual QDs via two-photon absorption. Key results are shown as Fig. 6. Insight into the process can be obtained by examining EM-CCD images collected with both infrared and green lasers on [Fig. 6(a)-i], with only the infrared laser on [Fig. 6(a)-ii] and with only the green laser on [Fig. 6(a)-iii]. We note that the infrared 1064 nm trapping beam is focused by the objective to a small spot (~1.2 μm diameter) while the green laser illuminates a broad area that is comparable to the field-of-view of Fig. 6(a). When both the green and trapping lasers are switched on [Fig. 6(a)-i], therefore, fluorescence can be seen both from an optically trapped QD around the nanoantenna gap (red circle) as well as a stuck QD to another part of the Si surface (yellow circle). When only the infrared laser is on, fluorescence is only observed from the optically trapped QD [Fig. 6(a)-ii], which we conclude to originate from TPA [32,36,44]. Similarly, when only the green laser is on, fluorescence is only observed from the stuck QD, as there is no longer a QD optically trapped around the nanoantenna gap [Fig. 6(a)-iii]. In Fig. 6(b), fluorescence emission of the optically trapped QD versus time is plotted, again obtained by integrating over a cross section covering 30×30 pixels from each EM-CCD frame. Both infrared and green lasers are on for 0 < t < 35 s, while only the infrared laser is on for t > 35 s. It can be seen that the QD blinking behavior occurs over the duration of the experiment. Emission spectra measured from the single optically-trapped QD when illumination is supplied by both lasers and by only the infrared laser are shown as Fig. 6(c). It can be seen that these have similar properties, peaking near the manufacturer’s target wavelength (655 nm), with the intensity being larger when both lasers are used. For comparison we include a spectrum measured when only the infrared laser is on and there is no QD trapped by the Si nanoantenna. This spectrum represents the background. In Fig. 6(d), we show the scatter plots of the tracked centroid locations of a single trapped QD. These data sets are collected with both two beams (trapping and green) and with only the trapping beam switched on. These are obtained with 1000 frames per data set over a time interval of 33 s and processed frame by
frame using a tracking algorithm [45]. These scatter plots quantify the QD localization by our all-Si nanoantenna tweezers. The corresponding histograms are plotted in Fig. 6(e), obtained with illumination from the trapping laser only [red dots in Fig. 6(d)], i.e., with two-photon-excited fluorescence. The full widths at half maximum (FWHMs) of Gaussian fitting profiles are 128 nm and 347 nm along the x- and y-axes, respectively. We conclude that the QD is trapped around the gap center, with the distribution including the combined effects of the optical gradient force and steric hindrance from the nanoantenna surface [8]. Our results demonstrate the optical tracking of a single QD trapped by an all-Si nanoantenna with nonlinear absorption, for the first time to our knowledge. In these experiments, because of the large TPA cross-section ($\sigma_{\text{TPA}}$), QDs can not only be excited via linear absorption from the green excitation laser (at wavelength 532 nm) by absorbing one photon, but also via TPA from the infrared trapping laser (at wavelength 1064 nm) by absorbing two photons simultaneously. We notice however that at the comparatively weak trapping laser irradiation level applied here [31–33] (~ 16.1 mW/μm²) and taking account of the electric field intensity enhancement produced by our all-Si nanoantenna (~ $5.39\times5.39 = 29$) [23], the nonlinear refractive index ($\gamma I_0$, where $\gamma$ is the nonlinear refractive index coefficient and $I_0$ is the light intensity) of CdSe is $\sim -6.9\times10^{-7}$ (<< $n_0 = 2.54$). This is extremely small, indicating insignificant modification to optical forces via the optical Kerr effect [42]. We note however that it has been reported that, in conventional optical tweezers with illumination from a femtosecond laser, the TPA process can enhance the trapping of QDs [30]. The use of femtosecond laser pulses with all-dielectric nanoantenna tweezers for the trapping of QDs and other nanomaterials might be an interesting topic for the future study. We also suggest that it may be fruitful to investigate the effect of the electric double layer coating on nanoparticles on trapping with optical nanotweezers, as recent work has shown it to be important with conventional optical tweezers [46].
5. Conclusion

In conclusion, we demonstrate that a single streptavidin-coated CdSe/ZnS quantum dot can be optically trapped by an all-Si nanoantenna and excited by the trapping beam via two-photon absorption. Our all-Si nanotweezers yield subwavelength light field confinement and enhancement with generating negligible local heating, which preserves the intrinsic properties of an individual QD and prevents photothermal effects including quenching and bleaching. We perform the tracking observation of the location and light emission of single trapped QDs by our all-Si nanoantenna, for the first time to our knowledge. The fluorescence behavior is visualized by an EM-CCD camera and analyzed with a spectrometer. Numerical simulations are also presented to calculate the optical forces applied to a single QD with the Maxwell stress tensor method. Future work could involve extending this platform by demonstrating the trapping of other nanomaterials such as single biological molecules.

Appendix

Optical trapping of single QDs

Single QDs can be optically trapped by Si nanoantennas via optical forces. In Fig. 7(a), both infrared and green lasers are on for $0 < t < 10$ s. At the beginning ($t = 0$), the QD is trapped by
an Si nanoantenna. This QD is only trapped for $0 < t < 4$ s and it is no longer trapped when $t > 4$ s. In Fig. 7(b), the green laser is switched on for $0 < t < 25$ s, and a single QD has been trapped at the beginning ($t = 0$). It can be seen from Fig. 7(b) that the QD is released from an Si nanoantenna when the trapping laser is switched off at $t \sim 12$ s. When the trapping laser is switched on again at $t \sim 18$ s, the QD cannot be re-trapped again because it moves far from the nanoantenna hotspots. This behavior is consistent with the short-range nature of the optical forces. In Fig. 7(c), the trapping laser is switched on for $0 < t < 90$ s and switched off thereafter ($t > 90$ s). The green laser is switched on always except for the time interval of 60 s to 80 s. As shown in Fig. 7(c), when the QD moves close to an illuminated Si nanoantenna, it will experience optical forces and can be trapped, resulting in an abrupt jump in fluorescence. At $t \sim 60$ s, the green laser is switched off and the optically trapped QD is excited only by TPA, with the emitted intensity being decreased. Turning the green laser on again at $t \sim 80$ s results in the emitted intensity reverting to the previous level. Switching the trapping laser off at $t \sim 90$ s results in the quick release of the QD from the hotspots. In Fig. 7(d), the trapping laser is switched on for $0 < t < 57$ s and switched off thereafter ($t > 57$ s). The green laser is switched on for $0 < t < 20$ s and $t > 45$ s and switched off for $20$ s $< t < 45$ s. Figure 7(d) shows the similar results as shown in Fig. 7(c), with the optical trapping by optical forces from $0 < t < 57$ s, being released after switching off the trapping laser at $t \sim 57$ s, and being excited by TPA only from the infrared (trapping) laser from $20 < t < 45$ s. The trap-and-release behavior confirms the predominant role of optical forces in the trapping process and that the QD is not attached to Si surface.

Fig. 7. Fluorescent intensity counts vs. time for trapping of a single QD. (a) $I_0 = 16.1$ mW/µm$^2$, 30×30 pixel cross section. (b) $I_0 = 17.3$ mW/µm$^2$, 35×35 pixel cross section. (c) $I_0 = 17$ mW/µm$^2$, 35×35 pixel cross section. (d) $I_0 = 12.7$ mW/µm$^2$, 30×30 pixel cross section. For (b)-(d), switching off the trapping laser results in the quick release of the QDs.
Trapping and sticking of QDs

In Fig. 8, we illustrate an occasion in which a QD is optically trapped by an Si nanoantenna but then becomes stuck to it. At the beginning [Fig. 8(a)], the QD moves randomly. The QD is then optically trapped by the nanoantenna and becomes stuck to Si surface by other forces [Fig. 8(b)]. It remains stuck thereafter [Figs. 8(c) and 8(d)], even after the trapping laser is switched off [Fig. 8(e)]. To illustrate the background level, an EM-CCD frame captured when both the green and infrared (trapping) lasers are switched off is shown as Fig. 8(f).

Fig. 8. EM-CCD frames showing optical trapping and sticking of a QD. (a) In this frame, QD is diffusing freely. (b)–(f) QD becomes stuck to Si surface. Purple circles in panels (d) and (e) indicate another suspended QD that moves freely by Brownian diffusion. The intensity of the infrared laser (1064 nm) is decreased in panel (d) [for (a)–(c): $I_0 = 17.3 \text{ mW}/\mu\text{m}^2$; for (d): $I_0 = 12.7 \text{ mW}/\mu\text{m}^2$]. Images are captured with auto-scaling of contrast, as provided by the EM-CCD camera and software. Scale bar: 8 µm.

Fig. 9. Tracking of fluorescent emission with green laser only from a single non-trapped free moving QD in solution as a function of time (35×35 pixel cross section).

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Author/s:
Xu, Z; Crozier, KB

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