**Low-noise Plasmonic Nanopore Biosensors for Single Molecule Detection at Elevated Temperatures**

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**Abstract**

Advanced single molecular analysis is a key stepping stone for the rapid sensing and characterisation of biomolecules. This will only be made possible through the implementation of versatile platforms, with high sensitivities and the precise control of experimental conditions. The presented work details an advancement of this technology, through the development of low-noise Pyrex/silicon nitride/gold nanopore platforms. The nanopores are surrounded by a plasmonic bullseye structure, and provides targeted and controllable heating *via* laser irradiation, which is directed toward the centre of the pore. The novel device architecture is investigated using multi-wavelength laser heating experiments, and are demonstrated to detect individual DNA molecules, under heated conditions. The plasmonic features, optimised through numerical simulations, are tuned to the wavelength of incident light, ensuring a platform which provides substantial heating with high signal-to-noise.

**Keywords**; Field Enhancement; Nanoplasmonics; Nanopore; Plasmonics; Temperature Control; Single Molecule Detection

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The development of devices aimed at the detection and analysis of biomolecules is an extensive area of focus for many research groups around the world.1–4 The forefront areas of this work demonstrate the enormous potential for this technology across a multitude of application areas.5,6 Nanopores are a class of biosensor which utilise nanometre-size holes to detect and characterise single molecules, without the requirement for molecular labels. Analyte molecules are effectively detected as they pass (translocate) through the nanopore, by observing the resultant experimental response.7 A growing area of research involves the perturbation of analytes, and local nanopore environment, during translocation. This interaction, either chemical or environmental, produces a response in the experimental observations – a means toward providing greater information about the analyte molecule. This approach incorporates; surface functionalisation, tuning surface chemistry, in addition to nanopore heating.8–11 The drive toward effective single molecule sensing is accompanied by additional techniques, used to observe translocation events, including; fluorescence imaging, tunnelling currents and surface enhanced Raman spectroscopy.4,12,13

Electrical sensing is the foremost approach in the detection of nanopore translocation events. This is usually facilitated by the use of two electrodes, inserted into two isolated electrolyte chambers, which are connected only *via* the nanopore.14 Upon the application of electrical potential, current flows, with the nanopore acting as the main source of electrical resistance. This means that any changes to the local nanopore environment (detection volume) results in a change in the current flow through the nanopore. This is exemplified by translocation events, whereby biomolecules travel through the nanopore resulting in either a current blockade, or current spike, depending on the molecules charge and the electrolyte used.15 Heating at nanopores has been reported in the literature, whereby additional heat increases nanopore conductance.16 This has also been shown to vary the behaviour of analytes as they translocate, providing an insight into the behaviour of single molecules.17–19

Nanopore heating can be achieved *via* two main methods; generalised heating of the nanopore membrane and surrounding environment, or more directly targeted heating using additional surface features.11,16,20–23 Precise nanopore heating using lasers can be attained by tuning the architecture of nanopores, and the surrounding material, in order to efficiently couple to incoming radiation. The most successful examples require structures made up of plasmonic materials that can drive radiation toward the nanopore, this provides a much greater efficiency relative to the use of non-plasmonic materials. These structures include; nanoparticles, bow ties and bullseyes, these have been shown able to be optimised to specific laser wavelengths.11,16,24 Bullseye structures provide a flexible platform, as the polarisation of the incident light does not have to be considered, allowing for uniform heating at any polarisation. The plasmonic bullseyes act to absorb and propagate radiation to the centre, directing heating to the nanopore.11

Crick *et al.* previously reported a plasmonic nanopore bullseye platform, which demonstrated precise and targeted nanopore heating.11 The major limitations of these devices originated from the device architecture (Au/SiNx on Silicon), and the large nanopore size (Ø - 80 nm). This provided relatively high electrical noise and a low detection sensitivity, rendering them ineffective as biosensors, but an excellent template for targeted nanopore heating. The devices used in this study are Pyrex substrate-based silicon nitride (Py-SiNx) solid state nanopores, formed using novel fabrication steps, and aimed at low electrical noise.4 The free-standing membranes are made up of gold-coated silicon nitride, however this is thinner than in previously reported device. The devices are fabricated *via* a series of thin film depositions, lithography, and etching. Focused ion beam etching is then used to fabricate the arrayed plasmonic bullseye structure (Ga ion FIB), however a smaller nanopore (Ø - 20nm) was achieved using He ion FIB. Ga ion FIB can provide material milling at this resolution, however utilisation of He ion FIB was essential for delivering these small features on the delicate free-standing membranes. The enhancements exhibited by these new devices are as follows; (i) low electrical noise architecture (Pyrex platform) – reducing the root-mean-square values from 400 pA to 23 pA, under full laser illumination, (ii) thinner membranes to increase single molecule detection sensitivity, and (iii) smaller diameter nanopores engineered to provide high signal-to-noise data.11 The previously established plasmonic bullseye architecture, was further improved specifically for the Py-SiNx devices through COMSOL simulations, aimed at targeted heating directed toward the nanopore.

The bullseye structure, milled using Ga ion FIB, is used to guide electromagnetic (EM) energy toward the nanopore is shown in Figure 1B (v). The nanopore milled using He ion FIB is shown in Figure 1C. The purpose of using the bullseye structure is to drastically enhance the EM fields within the nanopore *via* surface plasmon polaritons (SPPs), which are excited when phase-matching conditions are met by the correct ring structure. The ring period has been shown as crucial for the SPP phase-matching, is estimated to be 518 nm for an excitation wavelength of 632.8 nm at normal incidence.11 Utilising gold as a material for fabricating plasmonic antenna was important for localised nanopore heating. Previously reported examples of gold in nanoplasmonics have shown that the material can not only act to generate heat, but it also rapidly carries this energy, away due to its high thermal conductivity.25,26 This ensures that the heat generated and directed toward the nanopore does not spread to the localised electrolyte, but instead is concentrated at the nanopore or is absorbed by the gold film.

Anticipating the exact effect of an increased nanopore temperature on the behaviour of the DNA molecules translocating through is multifaceted. Literature studies have shown that elevated temperatures generally increase the velocity of Brownian motion of the DNA molecules in solution, and go on to demonstrate that this can increase translocation velocity (a decrease in event duration). Other factors including; optical trapping of DNA molecules, and the specific molecular/surface charges, which can also affect the movement of DNA at thermal gradients to varying degrees.18,22,27 This means that not only a change in DNA velocity must be considered, but also the electrolyte used, DNA charge, nanopore surface and bulk material and method of heating all contribute to variation in translocation behaviour at elevated temperatures.

The reported development of Py-SiNx nanopore devices, with surrounding plasmonic bullseye structure, are demonstrated as versatile biosensors able to function over a range of temperatures. The device architecture, demonstrated previously for generating targeted heating, is shown to readily applicable to a range of further biosensor technologies. This platform is shown not only to provide accurate heating with very low electrical noise, but also to act as a sensor in the detection and investigation of single DNA molecule translocations. The behaviour of the DNA molecules (10 kbp) was used as a proof of principle experiment, demonstrating faster translocations at induced elevated temperatures.

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**Figure 1**. (**A**) Schematic of the multi-step Py-SiNx device fabrication. (**B**) 3D representation of the plasmonic bullseye/nanopore structure etched into the free-standing membrane, where; (i) gold layer (47 nm plus 3 nm titanium binding layer), (ii) silicon nitride layer (20 nm), (iii) bullseye ring (~80 nm), (iv) bullseye periodicity (518 nm), and (v) nanopore (oversize for visualisation, Ø – 20 nm). Full device specifications are in the experimental. (**C**) SEM/FIB images of the bullseye/nanopore structures respectively, milled into the gold side of the free-standing membrane. (**D**) Experimental setup schematic, showing the laser irradiation and electrical detection. The Py-SiNx devices are submerged in 100 mM KCl, and a potential is applied across the membrane, the lasers are focused onto the gold side of the free-standing membrane.

**Results and Discussion**

*Plasmonic Py-SiNx Device Fabrication*

The plasmonic Py-SiNx devices were fabricated using a sequence of chemical vapour deposition, selective materials etching and lithography (Figure 1A). The plasmonic bullseye feature was milled into the gold (50 nm thick) using Ga ion FIB. This process was optimised to remove most of the gold, leaving behind a few nanometres of conductive material on top of the free-standing SiNx membrane (Figure 2). Initial estimations for milling time were made using standardised etch rates for known ion beam exposure. However, sequences of test milling, followed by visual inspection using scanning electron microscopy (SEM) were required to establish the ideal bullseye milling conditions. Common observations included; over-milling, resulting in removal of all the underlying gold or even membrane perforation, or under-milling, resulting in undefined surface features not able to function as a plasmonic antennae. This conductive layer proved essential in allowing successful milling of the nanopore in the subsequent stages, acting to prevent charging in the He ion beam. Pseudo-cross section simulation of the Py-SiNx membranes showed that variation of this thin layer of gold greatly affects the plasmonic behaviour and subsequent nanopore heating (Supplementary information S1 – further details). Bullseye milling which removed all gold, milling down to the SiNx, resulted in poor visualisation and low milling quality using the He FIB (Figure 2A/B/C). It was found that a thin gold connection provided a milling resolution indistinguishable from that achieved on an unmilled membrane. A significant feature of the He FIB instrument (Carl Zeiss ORION NanoFab) was the atomically sharp tungsten probe used in generating He ions. Both the quality of the probe formation, and the precise alignment of the guiding aperture, determined the achievable beam current, and its stability. Therefore, each milling session required calibration of the milling times required for complete membrane perforation, and nanopore formation. However, this was typically between 20-30 s, at a beam current of (7-8 nA) (Figure 2D). The final bullseye/nanopore structure was imaged using the He FIB (Figure 1C). Helium FIB was essential for small nanopore milling, as it allowed precise milling at the lengths scales required (Ø < 20 nm).28 Some Ga FIB instruments are capable of milling features of this size, however the delicate free-standing membrane would not be compatible with this, as milling position optimisation may be required. A comparison of Ga and He FIB can be viewed in Figure 2E.

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**Figure 2**. Schematic showing; (**A**) a profile view of plasmonic structure milling *via* Ga FIB, (**B**) a profile of nanopore milling *via* He FIB, and (**C**) visualisation using He FIB imaging. (**A**/**B**) Free-standing membrane comprises of SiNx (purple), and gold (yellow). Variation in the extent of Ga FIB milling, and its effect on He FIB milling quality are shown, whereby; (**i**) substantial over-milling partially perforated SiNx membrane [*nanopore milling quality* – very poor], (**ii**) complete removal of gold layer [*nanopore milling quality* – slow/low resolution], and (**iii**) leaving a thin layer of gold [*nanopore milling quality* – analogous to unaltered membrane]. (**D**) An example of a He FIB dose response milling array, used to gauge ion beam exposure time for nanopore milling. Each point was milled into the gold side of the Py-SiNx devices, beside the free-standing membrane, using a beam current of 7.6 nA, and exposure times of (i) 5, (ii) 10, (iii) 20, (iv) 30, (v) 45, (vi) 60, (vii) 75, (viii) 90 seconds. (**E**) SEM image of a nanopore milled into a Py-SiNx free-standing membrane using a Ga FIB beam. The nanopore size (~ 80 nm) is the minimum achieved with the FIB instrument used. (inset) Shows a He FIB image of a nanopore (~20 nm) milled using the He FIB. Scale bars are shown.

*Nanopore Sizing*

Nanopore sizing was carried out by direct imaging using the FIB instrument after milling. This yielded a value of 20 nm (± 3 nm), using the optimised milling conditions (Figure 1C). However, in order to characterise this more accurately, ionic current measurements were carried out. Chronoamperometric traces were taken at various voltages using the milled Py-SiNx nanopore device mounted in the fluidic cell. The cell comprised of upper and lower electrolyte reservoirs, each filled with aqueous KCl (100 mM), 10 mM Tris and 1 mM EDTA (pH 8) buffer, separated by the Au/SiNx membrane (full details provided in Supplementary Information). The data was subsequently used to generate IV traces which allowed an estimate of the pore conductance. Nanopore conductance of 0.54 nS (± 0.04 nS) at a 100 mM KCl concentration were observed, and this gives an estimate of ~ 7 nm (calculation shown in Supplementary Information) pore size. The nanopore size observed using FIB imaging (~ 20 nm) is likely to be larger than the actual size, as the gold layer (which is primarily imaged) is more easily milled than the underlying SiNx. This is supported by the conductivity measurements which yield values lower than optically measure (~ 7 nm). Previously reported devices, were characterised in the same manner, and yielded a similar trend.11 Electron microscope images suggested a pore diameter of 80 nm, however analysis of the electrical conductivity data suggested a smaller ~50 nm diameter.

These estimates however make the assumption of a completely cylindrical nanopore, which is almost certainly not the case, given the two membrane materials etch at different rates.29 The actual pore diameter is likely to lies somewhere between that overserved in the FIB images and the estimates based on conductivity data.

*Calculating Electric Field Enhancement and Temperature Changes*

Numerical simulations were carried out to examine both the electric field enhancement, and the predicted temperature changes, upon laser irradiation. The simulations replicated the experimental conditions, by matching the nanopore materials and dimensions (detailed fully in the Supplementary Information).30–38 The entire nanopore assembly was simulated using COMSOL Multiphysics. The incident radiation is x-polarized and normally incident. The symmetry of the system causes the plasmonic nanopore to be polarization independent. The field enhancement, defined as |E|/E0, in the nanopore is monitored and plotted in Figure 3A. As can be seen, there is indeed a broad peak in field enhancement around 625 nm in the case where the bullseye structure, with 518 nm periodicity, is present. This is in stark contrast to the case where there is no bullseye structure, which shows a field enhancement of only one to two times around 625 nm. The field enhancement can also be visualised (Figure 3B), showing a maximisation in the nanopore itself.

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**Figure 3**. Simulated electric field and heating results. (**A**) Plots the change in electric field enhancement with wavelength of incident light, laser wavelengths are indicated. (**B**) Shows electric field enhancement maps generated using 632.8 nm light, using a 5 mW incident beam. Both (**i**) Top-down, with nanopore enlargement (inset) and (**ii**) side-on images are shown. (**C**) Plots the nanopore temperature change, using a 5 mW beam at the given wavelength, both with and without milled bullseye structure. (**D**) Shows the temperature map for 632.8 nm laser, 5 mW. (**E**) Shows the equivalent plot for 532 nm light. (**D**/**E**) *Temperature value range; Minimum – 6K, and Maximum 33.5K*.

*Experimental Nanopore Heating*

Nanopore heating experiments were carried out using three laser wavelengths; 532, 632.8 and 685 nm, using a custom-built microscope (described fully in the supplementary information). These were chosen to probe a range of heating mechanisms modelled in the numerical simulations, which predict a more localised heating when using laser radiation around 632.8 nm (Figure 3D/E). A plot of the simulated heating profile can be found in the Supplementary Information (S2). The wavelength dependent temperature change (Figure 3C), shows that the bullseye structure provides a greater amount of generalised heating (~8K average improvement over the 500-700 nm range). The simulations also show additional heating in the wavelength region of maximised electric field enhancement (~ 625 nm, Supplementary Information S1).

The experimental heating agreed well with the numerical predictions (Figure 4), providing the highest amount of heating for the 532 nm laser (+37 K, at 5 mW), the 632.8 nm laser providing moderate heating (+20 K, at 5 mW), and the 685 nm laser heating the least (+8 K, at 5 mW). Although the magnitude of heating of the 532 nm laser is observed to be higher than that of the 632.8nm, the level of electrical noise can also be seen to be exceptionally high using the 532 nm laser (Figure 4B). The electrical noise of the devices, without laser irradiation is relatively low for this type of device (25 pA ± 5 pA, with 300 mV voltage). Upon irradiation with 532 nm laser there is an immediate onset of periodic noise (full details provided in Supplementary Information S3), this increases to a maximum of 160 pA (± 52 pA), at 5.12 mW. It was found that the noise increased in amplitude as the laser power was increased, however no change in the frequency of this noise was observed. Irradiation with the 632 nm laser, provides minimal change in noise, with a slight increase from the base value to 35 pA (± 7 pA) at 5.70 mW of laser power. The 685 nm laser shows no significant change in noise levels, even when the nanopore is targeted with 5 mW laser power. This noise originates from the mechanism of heating, whereby the 532 nm laser stimulates the interband transitions of gold, heating the general area of laser irradiation evenly. In contrast, the 632.8 nm primarily acts through exciting the surface plasmons which then propagation toward the nanopore, where the heating is targeted toward the nanopore.11 This kind of heating is observed to provide much lower noise, giving a much cleaner current-time trace (Figure 4B), and presenting a higher potential for the observation of single molecule translocation events under high laser powers.

Previous plasmonic bullseye devices showed similar heating trends, whereby 532 nm lasers provided greatest heating, and 685 nm the lowest. Under irradiation with similar laser powers, these devices (Au thickness – 100nm, SiNx thickness – 100 nm) provided 114K, 47K, and 21K for the 532 nm, 632.8 nm, and 685 nm lasers respectively. These values are higher than those observed in the Py-SiNx devices, although this can be explained through their thinner total membrane thickness (80 nm decreased from 200 nm), resulting in a lesser interaction with the incident light. The ratios of temperature increase between laser wavelengths is approximately the same for both devices. However, the 532 nm laser provides less than the expected heating in the Py-SiNx devices, and may be caused by the Py-SiNx devices possessing half the thickness of gold.

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**Figure 4**. Heating for the Py-SiNx bullseye devices. (**A**) Shows the power dependent change in nanopore temperature, using calculations based upon the experimentally observed changes in nanopore conductivity (*points*), and simulated temperature change (*lines*). See supplementary information for full details. (**B**) Shows the increase in baseline noise of the current-time traces, for devices heated, using maximum laser power for the 532 nm (green) and 632.8 nm (red) lasers, at an applied potential of 450 mV. (**C**) Shows top-down simulated temperature maps in the cases where (i) all four rings, and (ii) the central two rings are illuminated, with the respective side-on maps illuminating (iii) all four rings, and (iv) two rings (simulated illumination at a wavelength of 632,8 nm and a laser power of 5 mW). *Temperature value range; Minimum – 4K, and Maximum 19K*. (**D**) Plots a comparison of the simulated and experimental heating results for four and two ring illumination, using the 532 nm and the 632.8 nm lasers.

*Heating Dependence on Ring Illumination*

The bullseye structure is specifically designed to function with all four rings illuminated, as this is shown provide the greatest plasmonic enhancement. This generalised bullseye illumination is designed to excite surface plasmons, direct their propagation toward the nanopore, where targeted heating can be provided. The results shown in the previous sections have the entire four ringed bullseye structures completely illuminated by the incident lasers. The diameter of the focused laser spot approximates to that of the outer ring of the bullseye. Changing the excitation optics to include a higher numerical aperture objective, narrows and intensifies the laser beam, such that only 2 rings of the structure are principally illuminated (Figure 4). A plot of the simulated heating profile can be found in the Supplementary Information (S2).

Nanopore heating using the narrower beam provided heat changes of 55 K, 27 K and 4 K, for the 532, 632.8 and 685 nm lasers at full laser power (~5 mW) – full details are provided in Figure 4. This represents a +47%, +35% and -37.5 % change, relative to the heating *via* illumination of the full bullseye structure, for the 532, 632.8 and 685 nm lasers respectively. Thus by intensifying the laser light, the 532 nm laser produces the highest increase in heating. This increase directly corresponds to the intensification of light absorption closer to the nanopore, as the 532 nm laser induces interband transitions in the gold layer, and does not interact plasmonically with the surface. The 632.8 nm laser rise is due mainly to the intensification the plasmonic interaction at the smallest rings of the bullseye illuminated with the intensified beam. The periodicity of the two most inner rings (which is optimised for this wavelength radiation), allows for more intense excitation of the surface plasmons, and thus higher heating. The 685 nm laser conversely induces a reduction in heating when the intensified beam is used. This can be rationalised, as the 685 nm radiation does not activate the gold interband transactions, and is also less effective in exciting the surface plasmons compare to 632.8 nm radiation, as a result lower heating is achieved.

*DNA Translocations*

The previous sections detail fabrication and testing of the Py-SiNx devices, which demonstrates the enhanced device response gained through specifically engineered architectures. The higher sensitivity of these devices allows for single molecule detection. The following is preliminary data demonstrating the function of Py-SiNx devices as viable biosensors.

All translocation data relates to 10 kbp DNA in a solution of KCl (100 mM) with 10 mM Tris and 1 mM EDTA (pH 8) buffer. The DNA was diluted to a final concentration of 1 nM in the buffered solution and filtered using a 0.2 μm filter. The translocation of DNA through unheated nanopores was fully characterised, through thorough analysis of current-time traces (Figure 5A). A range of applied potentials were used to probe the DNA translocations, however the data presented is at a potential of 450 mV. The most important features of these translocation events are the dwell time and the peak amplitude, as they give an indication of molecular size and charge. It was found that the signal to noise for the events was excellent, providing events with peak amplitudes of ~170 pA with a baseline current of ~400 pA, and yielded an average 0.67 ms dwell time. Comparison to literature values can be made by approximating the translocation speed. The 10 kbp DNA has an average contour length of 3.4 μm, this provides an estimated speed of 5.1 mm/s, which compares well to previously literature values. Indeed, similar Py-SiNx devices have used 5 kbp DNA, and with have the contour length of 10 kbp DNA, the recorded dwell times were approximately half that observed in this study.4,39

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**Figure 5**. Translocation behaviour of 10 kbp DNA, using laser radiation with a 632.8 nm wavelength and applied potential of 450 mV. (**A**) Shows current-time traces where the spikes indicate DNA translocation events with full laser power applied - (i) 632.8 nm, (ii) 532 nm. (**B**) Shows the relationship between the average dwell time and the applied laser power, with this decreasing from 0.67 ms without laser applied, to 0.51 ms when the power of the laser is 5.70 mW. (**5B -** **Inset**) Shows typical DNA translocation events. (**C**) A 2D-histogram plot of peak amplitude against dwell time both without and with laser irradiation, clearly indicating the effect of laser irradiation on the translocation behaviour.

The effect of nanopore heating, on the DNA translocation behaviour was probed using the 532 nm, 632.8 nm and 685 nm lasers detailed previously, with the illumination of all four rings. It was immediately demonstrated that even though the translocation events show exceptional signal to noise without laser heating, periodic noise caused by the 532 nm laser visibly masked the translocations events (Figure 5A). This made the analysis of this data very challenging, as the noise albeit periodic had some variance in its frequency (see Supplementary Information, S3). Electronic filtering of the data was unable to remove this noise without greatly distorting the translocation events. The 632.8 nm laser heating provided minimal noise (as shown in Figure 4), and allowed clear visualisation of the DNA translocation events (Figure 5B). A change in translocation behaviour was observed as a greater laser power was applied, showing a decrease in the average translocation event dwell time. The dwell times were reduced from the original 0.67 ms, to 0.628, 0.562, 0.511 and 0.509 ms, as 10%, 50%, 80% and 100% laser power was applied. There was no substantial difference in the average peak amplitude of the detected events, with this remaining around 170-180 pA. There was no observed change in translocation behaviour when heated with the 685 nm laser.

Although the 532 nm laser gives the greatest magnitude of heating, the electrical noise introduced, even at low laser powers, means that translocation events cannot be monitored using an electrical detection method. The 685 nm laser provides no change in electrical noise level, compared to baseline levels, however the only a low level of heating is attained using a power of 5 mW (ΔT = +8K). This temperature rise did not give rise to a significant change in the translocation behaviour of the DNA molecules. With the average event dwell times remaining similar to that of unheated events. The 632.8 nm laser provided ample heating (+20K, at 5 mW laser power), and has been shown to induce a variation in translocation behaviour (decreasing from 0.67 ms to 0.51 ms when fully heated).

Previous work carrying-out DNA translocations on heated nanopores focus on the variation of three main translocation properties; dwell time, event amplitude and frequency. The general trend for the translocation times, is a shortening as temperatures are increased.17,18,40 This is explained through an increased Brownian motion of DNA molecules in solution, this is then carried forward as a reduction in time spent within the nanopore at elevated temperatures. The event amplitude is generally seen to increase at higher temperatures, and relates directly to the decrease in dwell time, and the need to pass the same molecular charge in a shorter time span. The shorter events must have the same area underneath the current-time traces (integration), thus the amplitude of the events must also increase to fit this. These are both observed in this study, moving from 0.67 ms to a 0.51 ms dwell time, and 170 pA to a 180 pA event amplitude. The translocation frequency is more complex, and depends upon multiple factors, which not only include the increased motion of the molecules, but systematic properties including; electrolyte composition and local temperature gradients which may affect the thermostatic and electroosmotic behaviour of the DNA.18,27 In the experiments reported here, there was no change observed in the translocation frequency.

**Conclusions**

The purposely designed architecture of the Py-SiNx bullseye devices, have been demonstrated to have exceptionally low-noise, providing an extremely high signal-to-noise for single molecule detection. The optimisation of device features to provide heating through plasmonic resonance (using 632 nm light), provides substantial heating of the local nanopore environment, without inducing interference with electrical single molecule detection. Intense electrical noise can be seen using 532 nm light, which provides higher nanopore heating, however the electrical signals from single molecules are entirely masked. Explained through device heating *via* the excitation of the interband transitions in gold. Variation of temperature with the detection of DNA molecules proves the novel device architectures are able to successfully function as biosensors. These devices offer a key step towards more advanced single molecular analysis, providing a versatile architecture, able to provide a high sensitivity of detection, in addition to a measure environmental control.

**Supporting Information.**

Additional detail of experimental procedures and numerical calculations, and further experimental results.

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