Electronic Polymers and DNA Self-Assembled in Nanowire Transistors

Mahiar Hamedi, Anders Elfwing, Roger Gabrielsson, and Olle Inganäs*

In the past decade tremendous advances have been made in a the field of structural DNA nanotechnology,[1] where techniques such as DNA origami[2] now demonstrate the ability to design and self-assemble 3-dimensional structures in aqueous environments, into exact shapes having resolutions close to the molecular distance between DNA base pairs,[3] in molar amounts. DNA nanotechnology is thus the most mature and precise 3D nano self-assembly platform to date. Naturally, further developments of electro-optically active materials, which by design can partake in the aqueous self-assembly process of structural DNA to form functional nanostructures, hold great promises for many fields including optical antennas/plasmonics,[4] nano mechanics, and nano electronics.

The use of metallic or semiconducting properties with DNA nanostructures requires materials beyond DNA; a considerable literature report studies of DNA in both insulating, semiconducting, metallic or even superconducting forms, however no electronic devices based purely on DNA have been demonstrated.[5] In addition, the flexible DNA chain is not necessarily best combined with classical metals and semiconductors with much higher elastic modulus. Metal and semiconductor nanowires have persistence lengths much longer than that of DNA, and typically, metals come as nanoparticles.

Electronic/conducting polymers, CPs, are very promising materials for the purpose of molecular self-assembly, as they span the entire range from insulators-semiconductor-metals. They are inherently 1D in their electronic structure, but retain the flexibility of the polymer chain, and have to date been demonstrated for the fabrication of numerous nanodevices.[6]

Dr. M. Hamedi, A. Elfwing, Prof. O Inganäs
Biomolecular and Organic Electronics
Department of Physics
Chemistry and Biology (IFM)
Linköping University
SE-581 83, Linköping, Sweden
E-mail: mahiar@ifm.liu.se

R. Gabrielsson
Departments of Organic Chemistry
Department of Physics
Chemistry and Biology (IFM)
Linköping University
SE-581 83, Linköping, Sweden

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In recent years there has been rapid developments in the field of CP based biosensors, where a library of different water-soluble cationic CPs have been assembled in solution onto oppositely charged biomolecules.[7] Here a body of work has demonstrated the use of cationic polythiophenes as optical DNA probes,[8,9] however these polythiophene functionalized DNA structures do not exhibit electronic conduction and are thus not suitable for nano device self-assembly.

Instead the route of aqueous polymerization of intrinsically conducting polymers (ICPs), using DNA as polymerization template has so far been explored. Three families of ICPs: polyaniline,[10] polypyrrole,[11–13] and poly(3,4-ethylenedioxythiophene) (PEDOT)T[14,15] have been oxidatively polymerized, using different oxidizing agents, from their respective monomer units dispersed in water, with the DNA phosphate backbone template acting as a counterion. The ICP polymerization route has been used for the demonstration of devices including the use of polyaniline/DNA complex for chemical sensor applications by controlling degree of doping,[16] and the use of the p(EDOT-N)/DNA composite for supercapacitor electrodes.[15]

However these polymerization environments with oxidative agents, and mostly at low pH, have poor compatibility with biopolymers, and often result in chain cleavage, as well as incomplete coverage along DNA chains, which is disruptive for electrical function. Furthermore the polymerization route of DNA/ICP results mostly in nano networks rather than free-standing nano devices. In general polymerization methods that use DNA double strands as a templates are non-specific, and disruptive to DNAs powerful capability of exact molecular self-assembly. This route is thus not ideal as a route towards mass self-assembly of individually well defined nanodevices. Instead true aqueous self-assembly of ICPs in the same manner as demonstrated for non-conducting polythiophenes has been the most desired, but until now not achieved route towards creating electronically functional DNA nanodevices.

In this study the fully acidic form of PEDOT-S was used for the purpose of self-assembly onto DNA. We have previously shown that PEDOT-S is a short polymer that is self-doped with ≈1/3 of the sulfonate side groups acting as the self-doping sites (see Supporting Information (SI)). The remaining sulfonate groups contribute to a net anionic charge, and a water-soluble polymer, with an intrinsic bulk conductivity of around 30 S/cm.[17,18] It has been shown that PEDOT-S can bind to oppositely charged cationic amyloid

protein structures in water and form conducting nano fibrillar networks,[19] and it has also been shown to form hybrid structures with synthetic peptides, and gold nanoparticles.[20]

In the case of DNA, electrostatic repulsion in deionized water conditions, between the anionic sulfonate group of PEDOT-S and the anionic phosphate backbone of DNA, should prevent interactions between these two polymers. Theoretical prediction for two polyelectrolyte chains of same charge show repulsive interaction, which however can be overcome by introduction of counterions. These can be monovalent, and charge neutralize the charged polymer. For divalent cations a different role is also envisaged, as they are shown to be efficient bridges, and in particular Ca^{2+} forms "inner-sphere" complexes with the phosphate backbone of DNA, leading to high adsorption rates at relatively low divalent cation salt concentrations.[21,22] By counterion condensation on the DNA chain, labelled Manning condensation, the net charge is reduced and attractive interactions can be found. Inspired by these findings, we chose to study the interaction of PEDOT-S and the double stranded λDNA in different Ca^{2+} concentrations. The studies were performed in de-ionized water with pH values close to 7. This pH value was chosen because PEDOT-S has a pKa near 3.5 (see SI), and in a pH close to 7, association to the Ca^{2+} is favorable. Increasing to higher pH can result in cross-linking of PEDOT-S with Ca^{2+} and at pH above 8, the polymer can start to precipitate.

An optical method was used for studying the PEDOT-S DNA interaction, where first a bis-intercalating fluorescent dye (YOYO-1) was bound to a fraction of the λDNA (0.005 mg/mL) base pairs, followed by addition of PEDOT-S from water (in 2/1 EDOT-S/DNA-base molar ratio). At very short range, the interaction of the excitons in the dye with the polarons in the ICP will lead to fluorescence quenching.[23] Therefore, decreased fluorescence intensity can be used as an indirect measure of bound PEDOT-S/DNA states (**Figure 1**a).

We found that in deionized water indeed very little fluorescence quenching was observed, most probably due to the repulsion between the anionic groups on PEDOT-S and DNA respectively. However, a sharp increase in the quenching for increased concentration of Ca^{2+} was observed (Figure 1c,d), with maximum values for concentrations in the vicinity of 10 mM Ca^{2+}. These findings suggest that divalent-bridges indeed suppress coulombic repulsion and lead to complexation between PEDOT-S and DNA. With much higher concentrations of Ca^{2+} up to 100 mM, we observed a decrease in quenching. This occurs as the polyelectrolytes are now effectively cationic, and charge repulsion becomes dominant once more.

For an infinitely long polyelectrolyte and sufficiently high charge density, condensation of counterions in a thin shell will occur,[24] and the effective charge of the polyelectrolytes thus be reduced. For DNA, with 2 negative phosphate groups per repeating nucleotide, the charge density is ≈ 2e-/0.33 nm, and the charge density length $l_C \approx$ 0.165 nm, while for the much shorter PEDOT-S l_C ≈0.39 nm, considerably lower. Assuming polymer repeat distances from studies of PEDOT,[25] the Bjerrum length l_B is

$$1_B = e^2/ek_BT$$

For water e = 80 at room temperature, giving $l_B \approx$ 0.7 nm. This is higher than the charge density lengths l_C for the two polyelectrolytes (DNA and PEDOT-S), which therefore both allow counterion condensation according to the Manning criterium

$$(1 - 1_C/1_B) = (1 - 1/\xi),$$

where ξ is the fraction of polyelectrolyte charges on which counterions have condensed. For DNA in water ξ = 0.76 and for PEDOT-S ξ = 0.44, meaning that in DNA three out of four nucleotides are charge

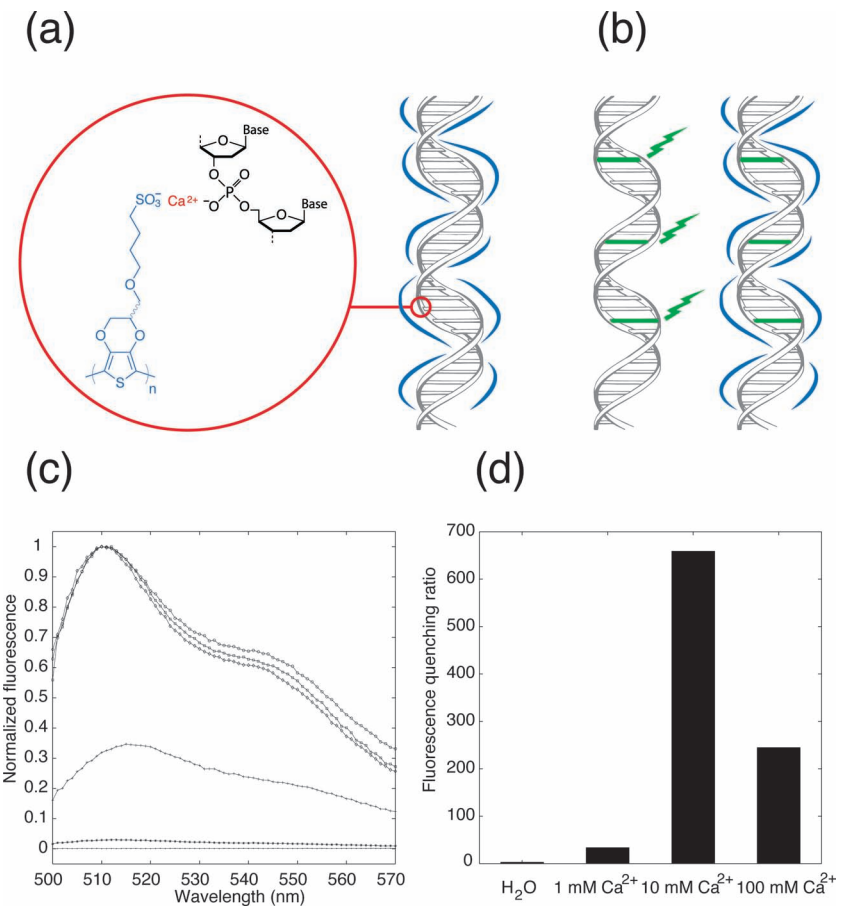

Figure 1. (a) Schematic picture of PEDOT-S/DNA molecular self-assembly with Ca^{2+} as a divalent cation bridge. (b) Schematic picture of fluorescence of the intercalated dye YOYO-1 (left) and the quenching of YOYO-1 by PEDOT-S binding (right). (c) Fluorescence spectra of λDNA/YOYO-1 complex in: (○) H_2O, (□) 1 mM Ca^{2+}, (◊) 10mM Ca^{2+}, and λDNA/YOYO-1/PEDOT-S complex in: (+) H_2O, (*) 1mM Ca^{2+}, (•) 10mM Ca^{2+} (d) Ratios between the fluorescence peak values of λDNA/YOYO-1 and λDNA/YOYO-1/PEDOT-S in de-ionized water and at different Ca^{2+} concentrations.

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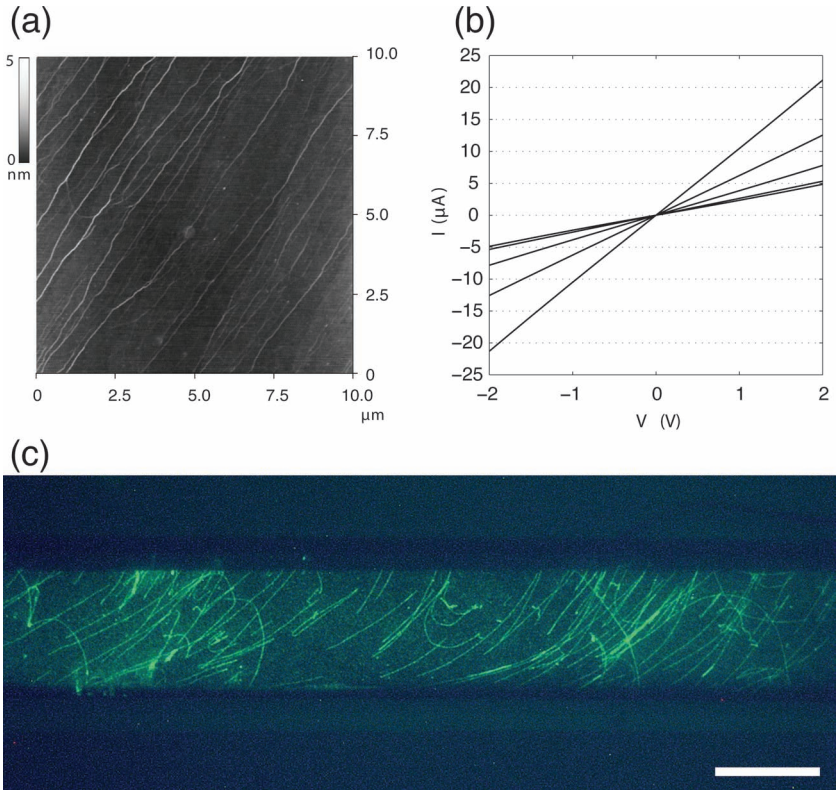

Figure 2. (a) AFM micrograph of λDNA/PEDOT-S (10 mM Ca^{2+}), stretched on Si substrate. (b) *I-V* curves of stretched λDNA/PEDOT-S, measured across the micro-electrodes on glass substrate, similar to (c). (c) Fluorescence microscopy pictures of λDNA/YOYO-1 (10 mM Ca^{2+}) stretched on glass substrate, across a gold micro-electrode gap. The black areas correspond to the gold electrodes as the fluorescence is quenched here due to metallic quenching. Scale bar 10 μm.

compensated and that PEDOT-S carries counterions at every second monomer. Note that the prediction for PEDOT-S should only be taken as a first guess; first, the chain is very short and arguments based on infinite chains carry little weight; secondly, this polymer chain is metallic and thus contributes a special electromagnetic shielding which is not included in the theory. The plausible reduction of charge on both polyelectrolyte chains will reduce coulombic repulsion, and with the divalent cations presumably also coordinate the two polyelectrolytes at a distance short enough to lead to efficient quenching of photoluminescence from the inserted YOYO emitter.

Comparing concentrations, we note that the 10 mM Ca^{2+} is higher than the concentration of both DNA nucleotides (15 μM) and PEDOT-S (60 μM with respect to monomers). If Manning condensation occurs, all available sites at both PEDOT-S and DNA could therefore easily be occupied, and overcompensation be the case, neglecting competition between ion condensation on DNA versus PEDOT-S. This will effectively cause an attractive interaction solely on coulombic mechanisms. In addition, experimental and theoretical studies have recently shown the existence of specific hydrogen bonds between the 3,4-(ethylenedioxy)thiophene (EDOT) and guanine,[26] and it is conceivable that such strong bonds form in the PEDOT-S/DNA complex once the charge screening and the cationic bridge allows the molecules to come in close proximity. It is further noteworthy that the optimum concentration of 10 mM Ca^{2+} is not higher than the concentration used in standard DNA buffers.

The fortuitous combination of effective Ca^{2+} charge screening at non disruptive concentrations and the bonding match between EDOT and DNA bases, seem to result in true aqueous self-assembly of a functional polymer on top of DNA, which naturally allow for further manipulation and self-assembly steps. We showed this by further deploying molecular combing of the PEDOT-S/λDNA complex (see experimental section). Molecular combing could successfully be performed on a number of different substrates, and the complex demonstrated behaviour very similar to bare λDNA in 10 mM Ca^{2+}. The retained combing capabilities are likely to be caused by the polymeric nature of PEDOT-S, similar to earlier demonstrations with fluorescent conjugated cations complexes with λDNA.[27]

AFM measurement of stretched PEDOT-S/λDNA (**Figure 2**a) showed a smooth DNA surface coating. This suggests that the binding sites where PEDOT-S could bind were saturated, since the concentration of PEDOT-S was designed to match the molar ratio of DNA binding sites. Minor inter-chain branching could be seen between the stretched λDNA wires, and this could be attributed to polymer chains bridging between few phosphate backbones.

The successful combing of the individual PEDOT-S/λDNA nanowires, where each wire could be elongated to over 15 μm lengths, allowed for electrical measurement by stretching the nanowires across gold electrodes with 15 μm gap spacing. Figure 2c shows a fluorescent picture of a YOYO-1/λDNA stretched across a 15 μm gap, acting as a pictorial proxy for the PEDOT-S/λDNA structures used in the electrical measurement.

I-V measurements in ambient conditions showed clear Ohmic behaviour. Figure 2b shows *I-V* curves measured for different samples made with the same combing procedure. The conduction further strengthens the proof that the λDNA is continuously coated with the ICP, PEDOT-S, to an extent, which allows for conduction throughout the extremely high aspect ratio nanowires, even after being subjected to the stress during molecular combing. A rough estimation of the conductivity and density of the conducting DNA nanowires (see SI), results in an average density of around 1 nanowire/ μm, which is close to the measured AFM and fluorescence pictures (Figure 2a,c). Although these estimations are very rough, they suggest that the majority of the nanowires are conducting and that the material conductivity of the complex is close to that of bulk PEDOT-S.

In order to test the conduction dynamics of the λDNA/PEDOT-S nanowires, we set out to construct an organic

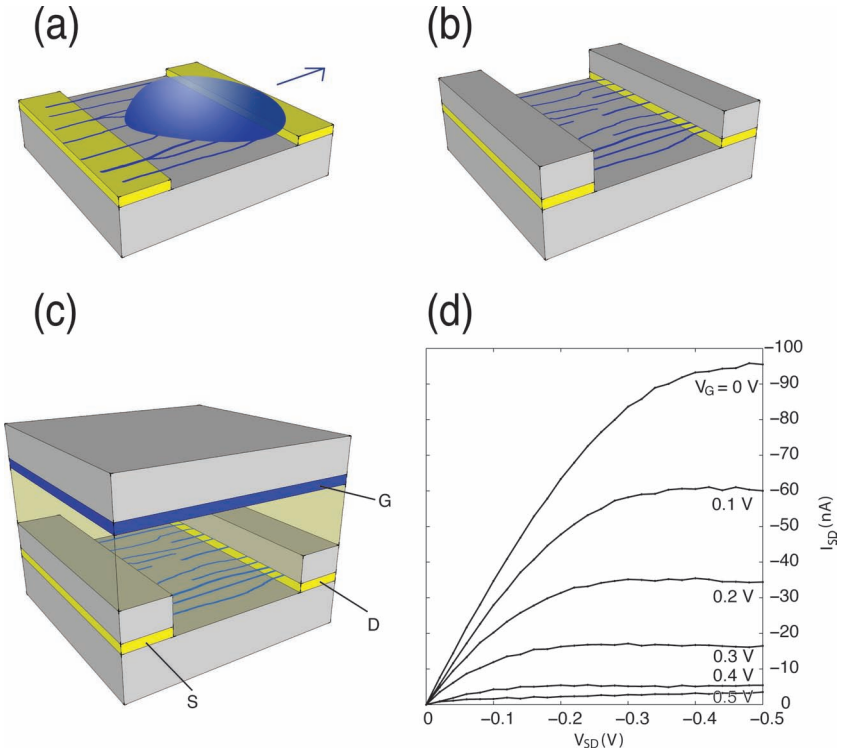

Figure 3. Schematics steps of the construction of the DNA nanowire transistor: (a) Stretching of λDNA/PEDOT-S nanowires across microelectrodes S,D (b) Addition of an electrolyte insulation barrier on top of the electrodes (c) Addition a PEDOT:PSS film acting as a gate G with an ionic liquid gel electrolyte in-between the channel and the gate. (d) Transistor transfer characteristics, of the device schematically depicted in (c).

electrochemical transistor, OECT, by micro mechanical assembly of additional components on top of the nanowires stretched across an electrode gap. **Figure 3**a–c show schematics of the assembly procedure, and the structure of the DNA nanowire OECT. Here the transistor channel comprises the collection of all single λDNA/PEDOT-S nanowires probed via the two gold pads (source, drain), and the channel conductance is modulated by changing the degree of doping in the ICP phase. The modulation is accomplished by reversible electrochemical redox reactions through an electrolyte that connects the channel to a working electrode, acting as a gate. We chose a uniform film of PEDOT:PSS as gate material and laminated this on top of the channel. The operating voltages of the device is a function of the low electrochemical redox potentials of the ICP, and the OECT operates as a p-channel, depletion-mode device in the third-quadrant, as PEDOT-S is oxidized, and polaron hole conducting in its neutral "on-state".[28]

An ionic liquid gel was prepared and used as electrolyte, primarily for the purposes of minimizing water, to circumvent possible disassembly of the λDNA/PEDOT-S, and to have a solid material for the construction of a solid device. Furthermore, ionic liquids have high ionic concentrations at room temperature and are known to result in highly stable electrochemical devices with pi-conjugated polymers.[29]

Figure 3d shows a transfer curve for the DNA NW OECT. The primary finding here is that the DNA/PEDOT-S complex molecule not only has high conduction, but also the ability to modulate its intra chain conductance.

The transistor characteristics resembles previous results demonstrated on planar[28] and micro fiber[30] PEDOT OECT devices. It is however notable that the saturation regime occurs below 0.5 V_{DS}, and that a depletion of almost two orders of magnitude is accomplished already at 0.5 V_G. In the previously reported PEDOT OECT devices both saturation and depletion seem to happen at higher voltages of around 1 V. One explanation for dynamics at lower voltages, could be that the PEDOT-S layer in the nanowires are probably single molecule thin, which could result in different dynamics due to minimum bulk ionic diffusion.[31] The DNA/ICP complex could also shift the redox potential, leading to different operational potentials. The on/off ratios for these devices were however less than 100, and this value is primarily limited by the contact between electrolyte and S,D metal contact pads, resulting in leakage currents.

In conclusion, the demonstrated aqueous assembly of an ICP with DNA constitutes an important step in the field of DNA nanotechnology, as it turns the DNA molecule into a uniform and dynamic semiconductor-conductor hybrid in water, without noticeable disruption of the DNAs molecular self assembly properties, as demonstrated by the combing experiments. Although λDNA is used as a nanowire model here, these findings should be directly applicable to any other higher order DNA structure with double helixes, such as DNA origami nano structures.

The findings here also open possibilities for further exploration of how water soluble ICPs, can interact with ssDNA and further partake in the hybridization process, of ssDNA into dsDNA.

Previously DNA has been demonstrated to retain its base pair matching capabilities in the presence of fluorescent conjugated polyelectrolytes.[32] This ability combined with the inherent similarity between conducting polymers and DNA in their organic polymeric nature, show that further evolution of ICP material and DNA nanotechnology could in relatively near future allow for massively parallel self-assembly schemes, where both polymer conductors and semiconductors can be positioned in very exact 3D coordinates at high resolutions.

Experimental Section

Aqueous Self-Assembly and Optical Characterization of PEDOT-S/DNA: λDNA (New England Biolabs inc.) was diluted in MilliQ-water in 1,10, and 100 mM $CaCl_2$ respectively, to the concentration of 0.005 mg/mL λDNA. The diluted λDNA solution was further

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mixed with the bis-intercalating stain, YOYO-1 (Molecular probes, Life Technologies), resulting in a 15/1 DNA base-pair/ YOYO molar-ratio, corresponding to 0.5 μM YOYO. After 2 h of incubation at room temperature PEDOT-S (1 mg/mL) was added to the DNA/ YOYO solution and diluted down to 0.02 mg/mL PEDOT-S corresponding to a molar ratio of 2/1 EDOT-S unit/DNA-base unit. The final mixtures were further incubated for 2 h at room temperature before performing fluorescence measurements.

Fluorescence measurements were performed using a TECAN saphire2 plate reader in 100 μL black plates with transparent bottom, at room temperature, with excitation at 490 nm.

Stretching and Characterization of λDNA: Metallic micro gaps were created on different substrate materials, including glass, silicon, plastics and mica by using a 15 μm diameter VECTRAN liquid crystal polymer microfiber as a shadow mask, utilizing the excellent mechanical strength and small thickness of VECTRAN fibres. Gold was then evaporated in a thickness of 30 nm with an adhesion layer of titanium oxide, and the shadow mask fibres were removed.

In the case of glass, and silicon substrates, the surface was then modified with PDMS (Sylgard 184 silicone elastomer, Dow Corning Corp.) by placing a bare PDMS stamp on the substrate for 10 s in order to achieve a hydrophobic surface, necessary for DNA stretching.

For electrical devices and AFM studies, 5 μL droplets of the λDNA/PEDOT-S complex (0.005 mg/mL in 10 mM $CaCl_2$ prepared according to the described procedure) were placed on the substrate and then gently blown across the surface with a nitrogen flow, so that the drop moved across the micro gap, resulting in molecular combing in the direction of the moving droplet bridging the gap. The surface was then dipped gently in milliQ water and further dried in the same blow direction with nitrogen in order to remove any excess salt and other possible residues.

Optical characterization of the stretched DNA was made by stretching YOYO/λDNA complex (YOYO-1 2 μM/ λDNA 0.005 mg/mL in 10 mM $CaCl_2$) on PDMS modified glass. A Axiovert inverted fluorescence microscope equipped with a 100 × 1.4 oil immersion lens and a 40× long-range objective for a better overview, was used. A filter for excitation at 470 nm (Nr. 09, Zeiss) was used and images were recorded using a CCD camera.

Preparation of Ionic Liquid Gel: The ionic liquid, 1-ethyl-3-methyl imidazolium ethylsulfate EMIM-ES was mixed with 2-hydroxyethyl cellulose HEC (Sigma Aldrich) in a 96/4 EMIM-ES/HEC weight ratio. The mixture was mechanically stirred and then heated to 130°C for 2 h to get complete mixture. The gel solution was then cooled and used at room temperature.

Fabrication and Electrical Characterization of DNA Wires and Transistors: Monofilament VECTRAN liquid crystal polymer fibres with 15 μm diameter (Vectran fiber, Inc.) were aligned manually using a stereo microscope and placed on top of the micro gaps containing stretched PEDOTS/λDNA nanowires. Next a clear protective lacquer (Electrolube inc.) was sprayed on the surface to cover all the gold pads, except for the parts containing the DNA nanowires, which were masked by the Vectran fibre (see Figure 3b). The lacquer was cured at 50°C for 15 min and acted as an insulating barrier for minimizing electrolyte reactions with the gold pads. A small droplet of the ionic liquid gel was placed on the channel and a gate was laminated from the top using a substrate covered with a PEDOT-PSS film acting as a gate material (see Figure 3). The two gold pads (source and drain) and the PEDOT-S gate were connected to a Keithley 2400 parameter analyzer for transistor characterization. IV-measurements were performed prior to adding the ionic liquid gel and manufacturing the gate. All electrical measurements were performed at ambient conditions and at room temperature.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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