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Quantized Conductance Measurement System for Liquids and Application to DNA Solution

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ABSTRACT

In this study, a system for undergraduate and graduate laboratories was designed in order to investigate quantized conductivity behavior of solutions. The quantized conductivity of the DNA solution was examined by using the designed system. The quantized conductivity peculiarity of DNA solution was observed. The same quantized feature couldn't be reached after repeated experiments for buffer solution, mutated DNA solutions. It is concluded that it is possible differentiate similar kind of solution by measuring the quantized conductance behavior.

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INTRODUCTION

Nanoscale researches have gained a great interest in recent years. Moreover, there is a need of students and researchers for the understanding and developing new ways with research of nanoscale [1]. One of the interesting research subjects in nanoscale is the quantized conduction [2]. There are several simple experiments related to nanoscale conduction [3,4]. Quantized conductivity was shown with these very simple systems by using the formation of nano size contacts with mercury [3] or gold wire [4].

On the other hand, several researches were conducted to investigate electronic properties of DNA molecules which can be defined as a continuous stack of aromatic heterocycles, the base pairs, which extends down the helical axis [5-7]. In literature, there is large dispersion of DNA conductance values. The results of several experiments show that DNA could be conducting, insulating or semiconducting [8] depending on the DNA may be in [9] or ambient conditions [10], or on an inorganic substrate [11]. It is known that interpretation of experimental studies and the results attained on DNA conductivity can be difficult. Measurement of charge transport mechanism in DNA solution being complicated because of many factors such as electrode type, medium of DNA [12].

Our work has potential to contribute to the studies on electrical conductivity of DNA based solutions with the advantage of its simplicity. In our study, quantized conductivity measurements conducted for the solutions of DNA, and mutated DNA by developing the system that was presented by Tolley et.al. [4]. Most important innovation which comes with our system is its ability to investigate quantized conductivity of solutions by using a relatively simple setup.

MATERIALS AND METHODS

The designed system has three main parts namely flexible plate with gold wire in a cuvette, a translation stage and an electronic circuit with computer interface (figure 1). Firstly, 99.99%-pure and 100 μm diameter gold wire prepared on a flexible plate covered with insulator band. The middle point of the wire was fractured to control the breaking point. Because of the applied force from the translation stage's tip the curve of plate can be controlled. The wire can be broken or reconnected by controlling the bend of the plate. It can be seen without applied force in figure 2a and with applied force in figure 2b. As a result, it is possible to create nano contacts during this breaking or reconnecting operation at the fractured part of wire [4].

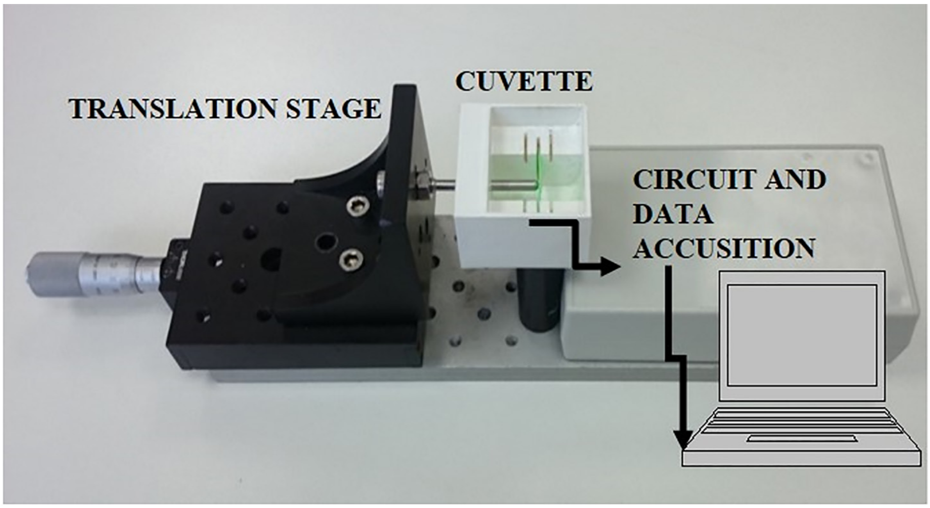

Figure 1. Quantized conduction measurement system.

Since the proposed system will be work in liquid medium, the gold wire was insulated to make sure that only fractured part will be in contact with the environment. The cuvette was printed with 3D printer to hold the flexible plate and liquid. After that, Thorlabs PT1/M 25 mm model translation stage extension connected to flexible plate. The cuvette system was sealed to prevent any liquid leakage during the measurement of solutions.

Resistance changes of gold wire were measured via the circuit presented in figure 3a. The circuit was designed to ensure that the voltage applied to DNA molecules in solutions is in millivolt range. For this reason, a resistor of 290 KΩ (R_ext) was connected in series to the resistance of 10 kΩ (R_p) and the gold wire. A 1.5 V battery (V_battery) was utilized to drive the circuit. Potential difference between the golden wire ends measured with DAQ, LabJack U3-LV unit connected to a computer. When the gold wire breaks, there will be no contact and the measured potential difference will be equal to the voltage on the 10 kΩ resistance. When the wire is broken, the micrometer could be turned to an opposite direction to provide reconnection. During both processes nano contacts occurs at the fractured point of the gold wire. The quantized conduction mechanism can be explained as a result of these nano sized atomic contacts..

Conduction or resistance values of gold wire can be calculated by using the quantized conduction equation $G_0(n)=(2e^2)/h\ n$ where n=1, 2, 3… is an integer value. The potential difference between the ends of the gold wire (Vlabjack) was formulated as in equation 1.

$$R_{gw} = \begin{cases} \text{Open circuit} \leftarrow \text{Broken} \\ 1/G_0(n) \leftarrow \text{Atomic contact} \\ \text{Short circuit} \leftarrow \text{Intact} \end{cases}$$

$$V_{labjack} = \begin{cases} \dfrac{V_{battery} R_p}{R_{ext} + R_p} = 54\ mV \leftarrow \text{Broken (open circuit)} \\ \dfrac{V_{battery}(R_p // R_{gw})}{R_{ext} + (R_p // R_{gw})} \leftarrow \text{Atomic contact} \\ 0\ V \leftarrow \text{Intact (short circuit)} \end{cases} \quad (1)$$

Where R_{gw} and $(R_p//R_{gw})$ is the resistance of the gold wire and equivalent resistance value of parallel connected resistances respectively. Calculated values of Vlabjack with respect to the quantum number n were plotted in figure 3b. According to the circuit described in figure 3, in the broken condition of gold wire the potential difference was calculated as 54 mV. As expected, the gold wire starts to behave like short circuit and the voltage value converges to the 0 V as the quantum number n increases.

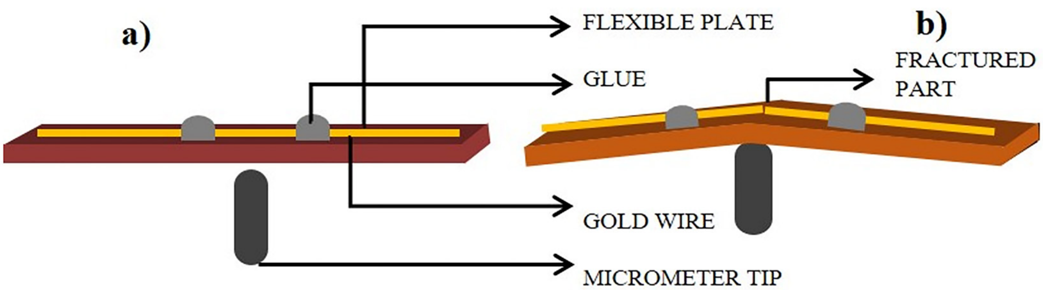

Figure 2. Flexible plates with gold wire (a) before bending (reconnected) and (b) after bending (broken).

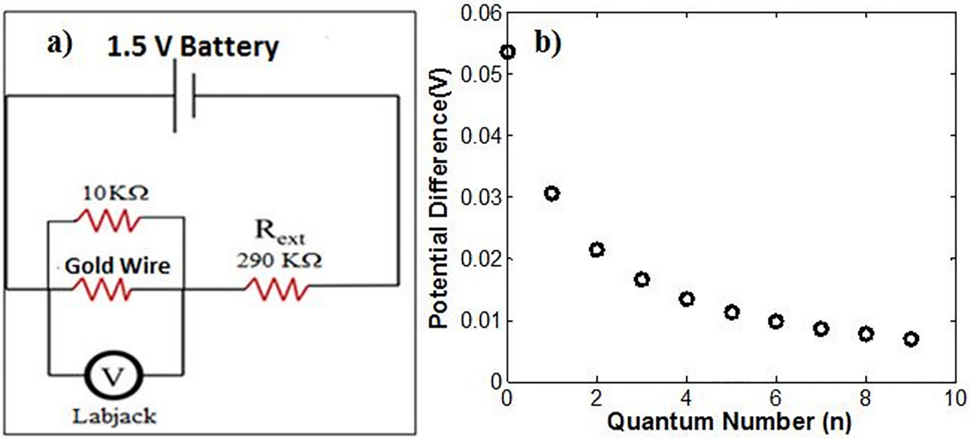

Figure 3. (a) The circuit diagram (b) calculated voltage values of $V_{labjack}$

Table 1. Solutions for liquid applications.

Analytes:	*Consantration of solution:*
DNA- Pure water solution	*2, 1, 0.5, 0.25 mg/mL*
DNA - Phosphor buffer solution (pH: 7.4)	*1 mg/mL*
Mismached DNA - Phosphor buffer solution (pH: 7.4)	*1 mg/mL*
Albumin – Pure Water	*1 mg/mL*
Brine solution	*1 mg/mL*
Acetic acid (liquid form)	*1 mg/mL*

Several solutions were prepared given in Table 1 to test the system. All solutions were prepared at the Ege University Science Faculty Department of Biochemistry. Herring sperm DNA (D3159) was purchased from Sigma Aldrich while the 14-mer DNA oligonucleotides as mutated DNA were obtained from Genset Oligos.

RESULTS AND DISCUSSION

Our initial measurements were taken without any liquid to ensure that our setup working as described in Tolley et.al. [4]. Measured levels of the quantized conductance were presented in the figure 4. Values up to 0.4 seconds were measured in the transition from broken state to contact state. The data given after 0.4 seconds was collected while the wire was breaking. Measured quantize levels were named according to the calculated values by using the equation 1.

Experiments with solution were conducted after the observation of quantized levels in control experiments. Wire left as an open circuit (broken state) just after the observation of quantized levels. This will ensure that distance between the broken wire ends at fractured point will be around nm range without any physical contact. DNA-distilled water solution of 1 mg/ml concentration was added to cuvette and voltage changes were observed.

Although there is no external mechanical effect, it is observed that the circuit gains conductivity after the DNA solution was poured into the system. Time over voltage graph of measurement was presented in figure 5. According to the resistance value of the circuit, depending on the quantum number n in which quantized voltage values can be seen. It is seen that voltage levels of the calculated and

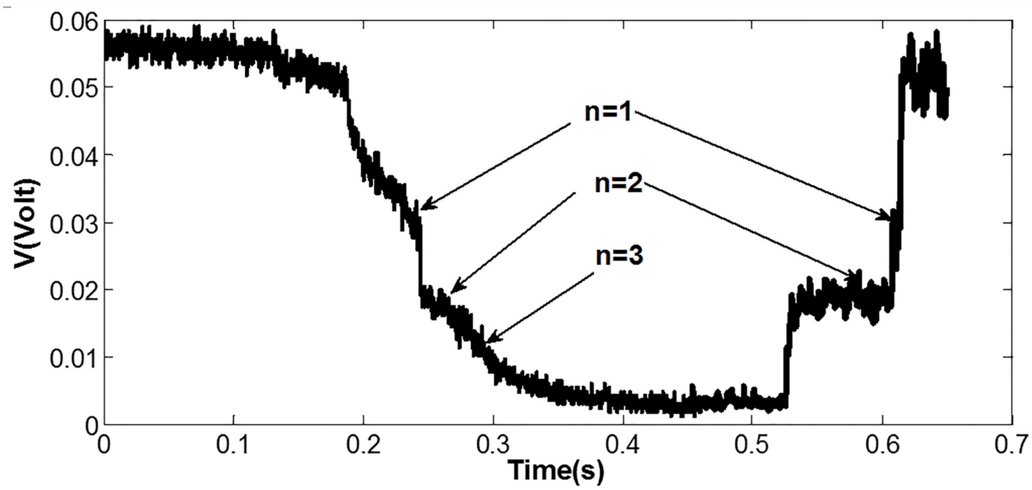

Figure 4. Measured voltage values while breaking the wire (0-0.4 s) and reconnecting the wire (after 0.4 s).

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experimental levels are approximately at the same level. The experiment was repeated after solving the DNA at phosphate buffer with 7.4 pH. The pH value of phosphate buffer solution was in the range of blood pH value (7.35-7.45). Similar quantized conduction result was observed again. Pure water and phosphate buffer added to the system separately to ensure that the effect is not caused by these solvents. For these solvents no conductivity change was observed.

Our results indicate that the observed conductivity change was clearly resulting of the DNA molecules in the solutions. Since DNA is negatively charged biomolecule, we believe that when it is added in to the medium, it connects electrically the two ends of the wire and make the wire short circuit. The voltage change before the first quantized level is not discrete as can be seen from the figure 5. We can explain this part of measurement with two factors. One of them is the impurities in the solution where they act as scattering centers and effects the quantized conductance levels [13]. Another one is the conductivity of solution. Before and during the formation of nano-channels the conductivity of solution effects equivalent resistance values in the circuit so the measured potential difference.

As a next step, different kind of molecules with strand structures were tried to further investigate the results observed. Prepared albumin solution, brine solution, and acetic acid solution were measured with the setup separately. It was observed that these materials have no effect on the conductivity. The results of other ionic solutions were ensured that the obtained quantized conduction result of DNA was not originated from the ions and other structures in the solution. As a final step to investigate the DNA conductivity, the mismatched DNA solved in phosphate buffer and it is added to the testing apparatus, and also for this case no conductivity change has been observed. It is known that the mismatched or single base bulged or protein binded DNA to not show any conductivity [5]. Electron

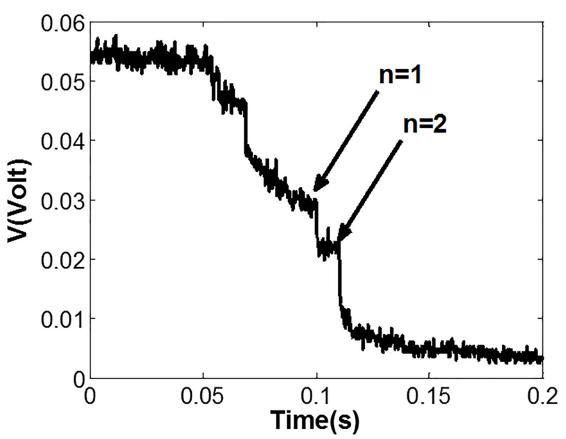

Figure 5. The potential difference change after 1 mg/mL DNA distilled water solution added into the system.

charge transport mechanism shown by these studies was in accordance with our results of DNA. So, it is detected that the quantized conductance change has been ensured by only healthy DNA solutions among the solutions presented in Table 1. Another important result was only DNA solutions were caused gold wire to switch open circuit to short circuit among the solutions tested.

Kasumov et al. observed that conductance of DNA value is in order of on the order of the resistance quantum (h/2e2≈12.9 kΩ) [14]. But following studies showed that DNA conductance values are found to be between 10−5G0 and 10−2G0 [15]. Studies of Xu et al. showed that conductance of DNA in buffer solution gives quantized conduction values during the stretching of DNA between the scanning tunneling microscopes (STM) electrodes. According to studies of Xu et al. occurrence of quantized conductance levels were due to the increase of the number of DNA molecules in the junction [9]. Conductance values of DNA in these studies were not in accordance with our results taken by our setup for DNA solution except the studies of Kasumov [14]. One of the reasons of observed high conductivity in this study can be explained by applied voltage value to the ends of wire. The applied potential difference between fractured wires ends causes to positively and negatively charged electrodes. This potential difference can cause forces on DNA and so can create nano contacts. Another reason might be the interaction between tips of broken wire and negative charges in DNA causes the DNA to act like a metallic structure. This possibility is supported by the researches where they found that the DNA is conductive [16].

The results of this study show that the presented setup could be used for to investigate quantized conductance behavior of solutions and materials in liquid form. One of the key features of our study is the quantized conductance observation of DNA. Another one is the only healthy DNA solution causes broken gold wire to behave like short circuit. We could not observe this phenomenon with the other prepared solutions. These results clearly indicate that this system can be used for DNA sensor applications and used for to distinguish the healthy DNA.

CONCLUSION

In our study, quantized conductance was shown with a relatively simple system for the DNA solutions. Because of this DNA structures are negatively charged, we believe that DNA is making the circuit conductive by going into the junction. As a consequence of several experiments, no quantization conduction observed for pure water, phosphate solvents, albumin solution, brine solution, acetic acid solution and mismatched DNA solution. These results show that obtained quantized conduction is specific to the DNA solutions. Our proposed system could be used for to determine healthy DNA in the solutions. Consequently, it is shown that the designed system can be used for investigation of the conductivity of the molecules in solutions.

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