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## **Towards optical detection of a single electron**

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# 6

## **Seeking for single-electron signals**

The discovery of the DBT/DBN molecular system with its high sensitivity to the electric field and the fabrication of SETs have paved the way to the optical detection of a single electron. However, there are still many challenges to overcome before achieving this goal. In this chapter, these challenges and the efforts to detect one single electron is described.

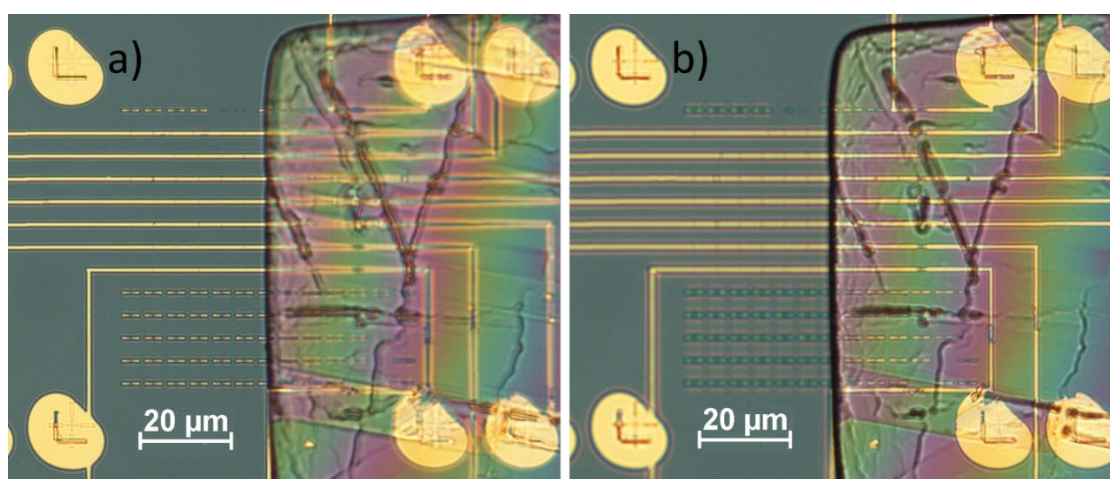
In chapter 4 we presented two methods based on using SETs to control and detect single charges. Although those methods require detailed knowledge of the electrical properties of the SETs (that are still under investigation), we performed optical measurements on our current SETs samples, which are reported in the present chapter.

## 6.1. Sample preparation

### 6.1.1. Crystal transfer to the chip

As we discussed in chapter 5, because of the high resistance and narrow insulator gaps of the junctions, the SETs are extremely sensitive to any potential change and to discharge of static electricity. Hence, the main and most important consideration in sample preparation is the protection of the sample against static charges. In all the steps from sample preparation to measurement, it is essential that sample, tools, and experimenter are properly grounded with the same ground.

The first step in preparing a sample is to transfer the DBT/DBN crystals to the chip. DBT molecules need to be placed within one hundred nanometers of the SETs to be able to sense the electric field of one electron. In chapter 2 we showed how DBT/DBN sublimated single crystal can be optically pasted on a glass surface. The same approach was used to attach the crystal to the chip and top of the SETs. Here, due to the presence of the nanostructures underneath the crystal, its contact surface with the sample is reduced and therefore its adhesion is reduced too. We also realized soon that imaging the SETs through the crystal is very challenging. This is because of the crystal anisotropy, of the difference in refractive index with the sample's environment and because of the thermal-change-induced cracks in the crystal which scatter the light. One solution to those problems is to use thinner crystals. Crystals with less thickness are more flexible and can bend between the nanostructure and better stick to the surface. Also, thinner crystals refract the light to a lesser extent, which improves the image quality. Further experiments have shown that crystals with a thickness of approximately 1 micrometre stick strongly enough to the surface.



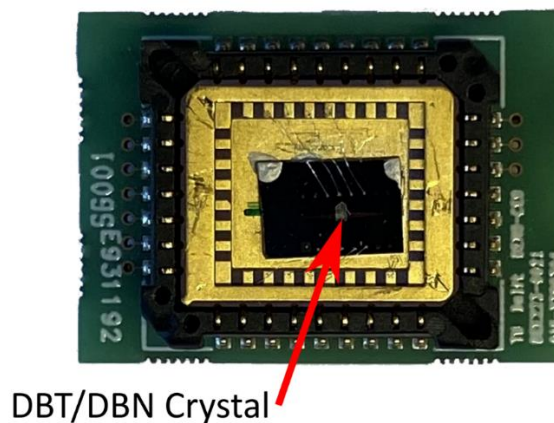
**Figure 6.1:** DBT/DBN crystal aligned and optically pasted on top of the SETs. a) Focused on the chip surface at the part without crystal. b) Focused on the nanostructure through the crystal.

The thickness of the crystals was measured before attaching them to the sample by a Broker DektakXT profilometer. The appropriate crystal was then transferred to the chip using steel

needles. Then, the crystal was adjusted on top of the SETs under an optical microscope. This step requires a lot of care and patience because any contact with lead electrodes can cause irreparable damage. Once the crystal was in place, it was gently pressed against the chip and stuck there. Figure 1.6 shows an example of a crystal successfully transferred to the chip. By comparing Figures 6.1.a and b, it is obvious that although the thickness of the crystal is only one micrometre, the difference in refractive index causes distortion and a large difference in focus.

### 6.1.2. Wire bonding

We used a wire bonder to connect the SETs to a chip holder and later to the power supply. A tiny droplet of silver paste was used to attach the chip to a chip holder attached to a printed circuit board. When the silver paste was dried and the chip was safely in place, all the pins on the circuit board were intermediately shorted from the back side and connected to same ground. This step was taken to prevent any potential change on the contact pad during wire bonding. A similar sample was used before wire bonding on the main sample to adjust wiring parameters such as power and time of ultrasound and applied force to the wire and contact pad. This significantly reduces the number of failed bonds. Since the contact pads are made of gold, we also used gold wire to have a stronger contact bond. The adhesion of wire bonding is often enhanced by heating up the sample, but this was ruled out in our case due to the presence of crystals on the chip. The wiring was always started with making the first bond on the chip holder then the second bond on the contact pad of the chip. This prevented damage to the contact pads if the bonding parameters were not properly adjusted. One of the contact pad of the holder was wired to the metal plate that the chip was pasted on it. This connection is the back gate and was used to manipulate the island potential. Therefore, Figure 6.2 shows the wire-bonded chip placed on the chip holder. The DBT/DBN crystal can be seen on top.

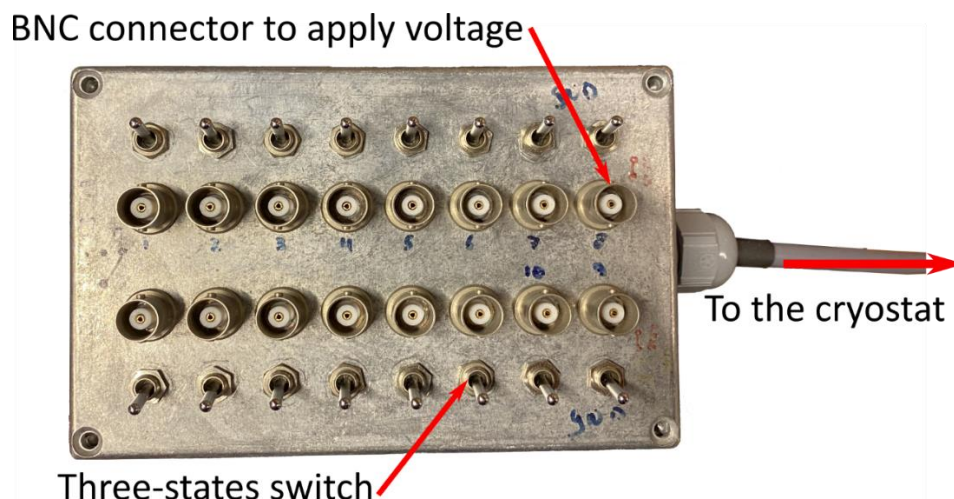


**Figure 6.2:** The fabricated chip wire bonded to the contact pads of the chip holder and held in place with conductive silver paste.

### 6.1.3. sample transportation

SETs are at more risk when they are connected to the big contact pads of the chip holder. To avoid damage during transportation, the chip holder was installed on the cryostat insert right after wiring and before disconnecting the intermediate shorts. The insert electrical contact pins are connected to a switch box showed in Figure 6.3. The switch box is designed to keep all the

pins at the same potential. Each individual key on the switch box has three states: ‘ground’ that internally connects the pins so they will be at the same potential, ‘off’ that disconnects the pins, and ‘measure’ that connects each pin to its related BNC connection. The BNC connections are used to apply voltages on the SETs electrodes. The switch box keys are kept on ‘ground’ during the preparation and cooling down. Also, all the other electrical connections such as temperature and pressure sensors, flow level meters for liquid helium and nitrogen, and vacuum pump were disconnected when the sample was inside the cryostat.



**Figure 6.3:** Home-made switch box to control SETs. The three-state switches are to ground, disconnect, and connect SET electrodes to the power supply.

## 6.2. Experimental section

The experiments were done at 1.2 K in a Janis flow cryostat. A tunable M-squared Ti:Sapphire equipped with TraScan unit and a WS6-200 HighFinesse wavemeter was used to excite DBT molecules at 756.6 nm, the wavelength of the ZPL of DBT/DBN (chapter 2). This laser system allows us to do excitation spectroscopy on molecules with sub-MHz resolution. A typical confocal microscope with a cryogenic objective (NA=0.8) was used for imaging. The same detector and sets of long-pass filters used in chapter 2 were used for detection of the fluorescence light.

Initially, a Keithley source-meter model 2450SMU was used to apply voltages on SET electrodes. However, we have found out that this Keithley device produces short and strong voltage pulses, especially when the voltage range is changed. These short pulses will unavoidably damage the SETs. To avoid damage caused by potential changes of commercial power supplies, a home-made battery-fed power supply was built and used to apply voltages. The power supply is shown in Figure 6.4. It is a very simple system with 4 batteries and two potentiometers that can provide up to 1.5 V on each of its 2 outputs with a resolution of 0.5 mV. The power supply outputs were connected to the switch box BNC connectors via short BNC cables (30 cm). Low bias voltages (0.5 mV) were applied to the source-drain electrodes and the gate voltage was scanned.

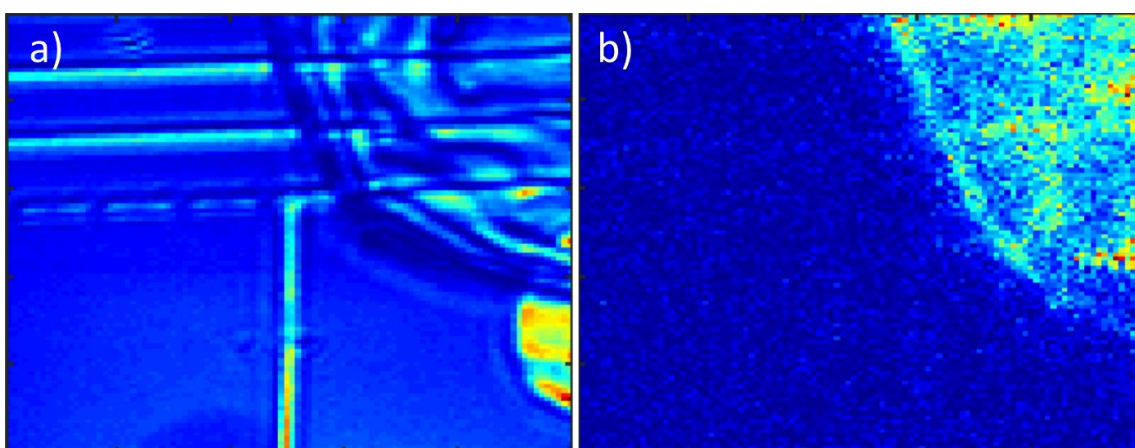




**Figure 6.4:** Home-made battery-fed power supply with two outputs and 0.5 mV resolution.

### 6.3. Results and Discussions

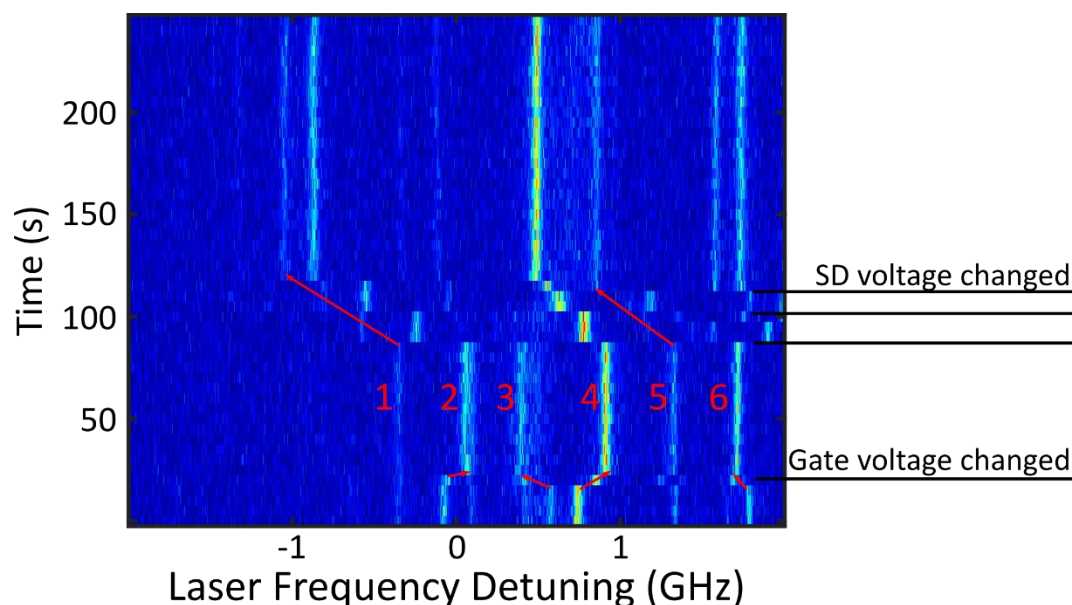
The biggest problem in optical measurements is determining the exact location of the SET islands. Figure 6.5 shows an example of sample images in reflection (6.5.a) and fluorescence (6.5.b) modes.



**Figure 6.5:** Confocal a) scattering and b) fluorescence images of the sample. The islands are not distinguishable in either case.

From these images, it is obvious that identifying the exact location of the islands is not easy. To feel the electric field of one electron, measurements must be done on molecules that are located a few hundred nanometres away from the island. Considering the size of islands (200 nm) and the imaging resolution that is about 400 nm, locating the islands is a serious challenge to the optical detection of an electron. Finally, the approximate location of the islands was determined by following the leads and the laser light was parked on the estimated position.

Another issue in optical measurements is finding molecules at the indicated spot. The TraScan unit brings the ability to scan the laser over large frequency ranges (THz scans). Applying this feature allowed us to find molecules at the desired spot. Figure 6.6 shows more than 6 molecules which were found at the estimated location of the island.

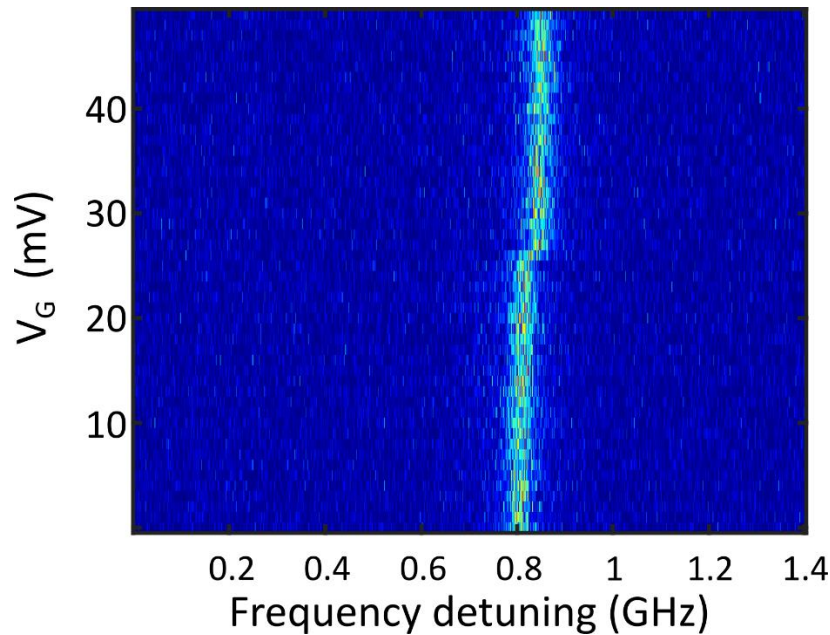


**Figure 6.6:** Molecule response to the Gate and Source-Drain electric field presents information about their approximate location.

First, the response of the molecule to the gate and source-drain (SD) voltages was investigated. The molecules' response to different applied electric fields gives information about their location. For convenience, the electric field caused by applied voltage to the gate and SD is called  $E_G$  and  $E_{SD}$  respectively. The first jump in Figure 6.6 is due to the  $E_G$  when all the other jumps (2<sup>nd</sup>, 3<sup>rd</sup> and 4<sup>th</sup>) are response to the  $E_{SD}$ . Molecules 2 and 4 were blue-shifted while molecules 3 and 6 were red-shifted with the  $E_G$ . That means their dipole moment is oriented differently with respect to the gate electric field. All 4 molecules were also shifted with  $E_{SD}$ , indicating that these molecules are placed close to the SD electrodes. Interestingly, molecule 1 and 5 did not show any shift under  $E_G$  while they jumped to the higher frequency under  $E_{SD}$ . Most probably these molecules are located on top of the island that the gate electric field is shielded by the island, but they were shifted by  $E_{SD}$ . Then by keeping the voltages constant, the molecules also remained stable.

In the case of molecules showed in Figure 6.6, many shifts were observed, generally due to the  $E_G$  and  $E_{SD}$ . This observation was also true in other measurements and molecules always showed a linear response to  $G_E$  and  $SD_E$ . In many cases, no distinct shift that could be specifically related to the presence of an electron in the molecule's vicinity could be observed or at least recognized. However, one case, shown in Figure 6.7, was an exception. In this measurement, again SD was in low bias and gate voltage was ramped up. The molecule does not show a large response to the  $E_G$  (the small shift is because of laser drift). This indicates that the molecule is on top of the island and does not feel  $E_G$ . Then suddenly at 25 mV for the gate voltage it jumped by 20 MHz (about one linewidth of ZPL). 25 mV is within the estimated size of a Coulomb diamond (50 mV) for the fabricated SETs. The jump can be a signal of adding one extra electron to the island and therefore, of the optical detection of one electron. But it may also have other reasons such as a sudden change in trapped charges distribution in the vicinity of the molecule (in the substrate, crystal, or aluminium oxide for instance) or charge leakage. Although there are compelling reasons to believe the observed jump is related to adding one extra electron to the island, without accurate knowledge of the electrical properties of SETs it is hard to make a definite statement about it.





**Figure 6.7:** A molecule placed on top of the island experience a sudden shift of 20 MHz at gate voltage around 25 mV. The observed jump could indicate the presence of an extra electron on the island and indicate the optical detection of a single electron. The scan time was 250 s and molecule was extremely stable during the scan.

#### 6.4. Conclusion

In Chapter 2, the optical properties of DBT/DBN system were examined and it was shown that the DBT molecules are very stable and show large linear Stark shift. In this chapter DBT/DBN crystals were used for the optical detection of a single electron. Sample preparation and crystal transfer to the chip were discussed. Tips and precautions related to sample transportation and measurement preparation were stated. Despite the difficulties in identifying the location of the island, by investigating the response of molecules to the electric  $G_E$  and  $SD_E$ , information about the approximate location of molecule with respect to the island was obtained. Finally, in one of the measurements, a signal that possibly indicated the presence of an electron in the vicinity of the molecule was observed. However, it is hard to claim the optical detection of a single electron due to the lack of electrical properties of SETs and repetition of the measurement. The most important step in following is measuring the electrical properties of SETs and investing in performing simultaneous optical and electrical measurements.