



Universiteit
Leiden
The Netherlands

Towards optical detection of a single electron

Moradi, A.

Citation

Moradi, A. (2021, February 23). *Towards optical detection of a single electron. Casimir PhD Series*. Retrieved from <https://hdl.handle.net/1887/3149275>

Version: Publisher's Version

License: [Licence agreement concerning inclusion of doctoral thesis in the Institutional Repository of the University of Leiden](#)

Downloaded from: <https://hdl.handle.net/1887/3149275>

Note: To cite this publication please use the final published version (if applicable).

Cover Page



Universiteit Leiden



The handle <https://hdl.handle.net/1887/3149275> holds various files of this Leiden University dissertation.

Author: Moradi, A.

Title: Towards optical detection of a single electron

Issue Date: 2021-02-23

Summary

Single-molecule spectroscopy has become a powerful method for using organic fluorescent molecules in numerous applications. Along with sensing applications in biology and solid-state physics or a variety of applications in quantum information technology, molecules offer interesting possibilities for fundamental research. One of the very interesting areas is the study of charge transport and electric field sensing at the nanoscale. Developing molecular nanosensors for electric fields can not only help to fundamentally explore the motion of charges in conductors and semiconductors but can also lead to very sensitive and accurate instruments for quasi-static charge tracing or even single-electron charge detection. Such research could eventually lead to the construction of precise electric field sensors that can act as an interface between the quantum state of an electron and the outside world. With this in mind, we developed fluorescence molecular systems and electronic circuits with the aim of electric-field sensing and optical detection of one single electron. Chapter 1 gives a general overview of cryogenic single-molecule spectroscopy and Stark effect.

In Chapter 2 of this thesis, I describe a very sensitive molecular probe for the electric field. The system is based on embedding dibenzoterrylene (DBT) as the probe (guest) molecule in a 2,3-dibromonaphthalene (DBN) host matrix. Single crystals of DBN doped with DBT were grown by co-sublimation. DBT molecules embed along the crystal a-axis by replacing 3 host molecules. The herringbone structure of the DBN crystal introduces distortion and bending in a DBT molecule and breaks its symmetry. The DBT deformation combined together with the effect of electronegativity of the bromine atoms, induce a large electric dipole. The dipole moment change of 2.1 D was measured experimentally, which is in good agreement with the calculated dipole moment change of 2.46 D. As a result of inducing such a dipole moment, a large and homogeneous linear Stark shift of up to 2 GHz/kVcm^{-1} was measured for a DBT molecule. For a field of 10 kV/cm, this shift is 600 times larger than the linewidth of the molecule's Zero-Phonon line (ZPL) and indicates its high sensitivity to the electric field. We also studied the optical properties of the system. A very stable narrow ZPL with a linewidth of about 40 MHz and brightness of about 10^7 emitted photons per second were measured experimentally. Quantum chemistry calculations confirmed our finding of a large dipole moment of the DBT molecule induced by the host matrix. Our simulations confirmed that the insertion of DBT into the matrix unit cell can be done in 8 different ways, corresponding to the 8 matrix DBN molecules in the unit cell. Although these sites lead to different orientations of dipole moments (and therefore to different Stark coefficients in an arbitrarily applied electric field), in terms of optical transition energy, they all have the same energy due to the 3 mirror symmetries of the system. This system can be used for electric field probing in nanostructures or as an excellent tuneable single-photon emitter. More importantly for our project, it is an excellent candidate for optical single-electron detection.

In Chapter 3 we studied a novel phenomenon and described its capability to tune the optical transition of fluorescent molecules. The light-induced charge carrier in several guest/host molecular systems was explored. Generated charge carriers in the vicinity of the molecule generate a local electric field and produce a Stark shift. These charges have a long lifetime of the order of several hours. The generated Stark shift can be used for long-lived, electrode-less, fine tuning of optical transitions in fluorescent molecules. We then introduce a photoionization model, to describe the charge generation and propagation in the matrix. The effect was used to tune ZPLs of DBT molecules in three different host matrixes – polycrystalline naphthalene, single crystals of 2,3-dibromonaphthalene and anthracene nanocrystals. For the nanocrystals, a shift of more than 100 GHz was obtained. Our collaborators in Florence used the effect for frequency matching of 5 different DBT molecules spatially separated from each other. This fine tuning offers many possibilities for applications of molecules in quantum technologies such as integrating many single-photon emitters in molecular electronics and photonic circuits.

In Chapter 4, I introduce single-electron transistors (SETs). The working principles of SETs are discussed theoretically, and their operation and properties are presented. The number of electrons inside a finite island is an integer number and can be controlled precisely by adding electrons one by one, if the Coulomb blockade condition is satisfied. This offers the idea of trapping a single electron in a metallic island in the vicinity of a molecular probe that is sensitive to electric fields, such as the DBT/DBN system. This integrated circuit can be employed to detect a single electron optically. In this hybrid approach the number of electrons inside the island can be controlled electrically. Simultaneously, the molecule can translate the existence of extra electrons into a shift of its optical transition, that is detectable optically. Although SETs exist for decades, their use for this application requires specific conditions. The essential aspect is that the electric field created by the charged island should not be shielded by the leads or by any other bulk metal. A suitable configuration of a SET for single charge detection was designed. It consists of a 200 nm island with very pointed leads that have minimum overlap with the islands. A COMSOL simulation shows that the electric-field strength of a trapped electron in such a SET device can be sensed up to 200 nm away from the island, considering DBT in DBN as the probe molecule.

Considering all the requirements and the facilities in our lab, a fabrication method was developed to manufacture the SETs. In Chapter 5, the different fabrication approaches used are presented. The effort started with e-beam lithography. Despite using several tricks such as overdosing the electrodes with respect to the island in a SET configuration, the poor resolution of e-beam lithography prevented the fabrication of the tunnelling gap. Another fabrication method based on self-assembly of gold nanoparticles and nanorods was developed. 40 nm gold nanospheres (as island) were assembled in between two nanorods (as electrodes) by using homocysteine molecules as the linker. The lack of dedicated fabrication facilities prevented us from following up this approach. Finally, a 10-step hybrid method was developed to fabricate the SETs. In this method the e-beam resolution limit was overcome by separating islands from electrodes with an insulating layer that acts as the tunnelling gap. In this method, 200 nm islands were patterned using

e-beam lithography, and then a tunnelling gap was grown with atomic resolution using atomic layer deposition. The ability to precisely control the thickness of the insulating layer allows us to create a broad distribution of tunnelling gaps. A large number of transistors were damaged during transportation and electrical measurement by static electricity and instability of the electrical potential in the electronic setup. After failing to measure the SETs in Leiden, we pursued the electrical measurements in collaboration with Professor H. van der Zant's group at Delft University of Technology.

In chapter 6, the efforts to detect one electron optically is described. The DBT:DBN crystal was optically pasted on the fabricated chip. The optical measurements were performed at 1.2K by immersing the sample in liquid helium. Due to the difference between the refractive index of the crystal (1.6) and the liquid helium (1.02) and to the formation of many cracks in the crystal during cool-down, finding the SETs underneath the crystal turned out problematic. The approximate location of the islands was determined using the matching of microscopic images and studying the molecular response to source-drain and gate electric field. Afterwards, source-drain were kept in low bias voltage and the gate voltage was scanned while excitation spectrum of the molecule was recorded. Gate voltage manipulates the potential of the island and allows one electron to enter the island at a certain voltage. Therefore, the electric field of the electron can be observed as a jump in the molecular spectrum or nonlinearity in the Stark shift. Although such a signal was observed, the reproducibility was not sufficient to claim optical detection of a single electron. The efforts to detect one electron optically are being continued.