Summer Project Report
Designing a resonator for molecular spintronics

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In August and September 2019, I carried out an eight week project in Prof. Arzhang Ardavan's group in the Condensed Matter sub-department of the University of Oxford Physics department. I received funding from the Physics department as well as free meals and accommodation from Merton College, and I am very grateful to both institutions for their support in allowing me to carry out this project.

One of Prof. Ardavan's current main research interests is in molecular magnets. Many atoms and molecules have a non-zero total electron magnetic moment (the total of the magnetic moments due to the intrinsic spins and due to the orbital angular momenta of all the electrons; a magnetic moment can be thought of as a microscopic bar magnet). In general, in the absence of any external forces, there is no preferred direction for the magnetic moment, which means that the direction of the magnetic moment of the molecule fluctuates on scales much faster than can be measured and averages to zero. This is in contrast to macroscopic objects, containing many many atoms, where a permanent magnetic moment can arise because the presence of the other spins in the system and interactions between them breaks the symmetry of the system. However, in certain molecules, there are two strongly preferred directions for the magnetic moment to lie in due to the geometry of the molecule and a large energy barrier for flipping between the two. This means that the molecule can have a spin and magnetic moment whose direction is constant for timescales on the order of microseconds, which is a long time compared to the time required for measurement. These molecules are interesting both from the point of view of pure science and because they have the potential for use in quantum computing.

In order to use molecular magnets for quantum computing, we need to be able to individually control the spin states of a large number of them. This is usually done using magnetic fields because magnetic fields can interact directly with magnetic moments. However, it is difficult to produce magnetic fields which are confined to a small area of space, whereas it is easy to produce localised electric fields. Therefore, the group is working on finding molecular magnets which respond to electric fields as well as magnetic ones. This is possible because the electric field can alter the geometry of the molecule in such a way that an internal magnetic field is produced. In addition, the timescale on which the magnetic moment of the molecular magnet randomly flips (the relaxation time) needs to be long (at least a few microseconds). This is more important with electric field driven transitions because the time taken to drive the transition is longer. The group has found a molecular magnet whose energy levels depend on an applied electric field and which has a relaxation time of about 8 microseconds for a particular transition with other useful properties. This system is therefore a good candidate for spin control by electric field.

We can investigate the state of electron magnetic moments using a technique called electron paramagnetic resonance (EPR). In EPR, electromagnetic waves in the microwave range are used to drive transitions between different angular momentum states of an atom or molecule with a non-zero total electron angular momentum, and so a non-zero electron magnetic moment. The sample is placed in a microwave resonator, which amplifies the microwaves which are at the desired frequency. Because most atoms and molecules respond to the magnetic field component of the microwaves, resonators are usually designed such that the sample is placed where the magnetic field is at a maximum and the electric field is at a minimum. However, we wanted to show that we could control the spin using the electric field component, so we needed to design a resonator where the sample would be placed in a maximum of electric field, and where the magnetic field would be as small as possible at the sample location, and where the electric field would be as uniform as possible at the sample location. In addition, the resonator needed to have the right resonant frequency for the transition we were interested in and it needed to fit inside the pre-existing equipment for cooling the sample (as a measurable net transition rate is only observed at liquid helium temperatures).

My project was to design a resonator which would satisfy all our constraints. My supervisor had already come up with an idea of a rough geometry but there were a large number of parameters which could be varied to optimise the design. In order to investigate the effects of changing the parameters, the electric and magnetic fields inside the resonator had to be simulated. This required us to numerically solve Maxwell's equations (which describe the electric and magnetic fields) inside the resonator, which we did using finite element analysis, which divides the space up into a discrete mesh on which the equations are solved. I put together some code to do this using several free, open source packages and I was able to verify the results using a piece of proprietary software. Putting the code together myself meant that I gained a much better understanding of both the underlying physics of resonant cavities and the mathematical methods used for the simulation than had I only used pre-existing software.

I then had to vary the parameters until I found a combination which led to the right resonant frequency, with the smallest possible magnetic field at the sample position, and with other modes of the cavity being separated in frequency by a long way from the desired mode. In addition, the design had to be sufficiently large to leave room for the sample and for the coupling to the source of microwaves, but sufficiently small to fit inside the cooling apparatus. We also wanted to be able to tune the resonant frequency of the cavity, which led to more design challenges as the cavity had to be sensitive to changing the total length, as this was the only parameter which it was physically possible to vary continuously.

I next had to put together a design for the workshop to make. This meant that as well as the dimensions of the interior of the cavity, I had to think about things like where all the screws would go to hold it together and had to deal with real world constraints such as the size of connectors which it is possible to buy. This was a side of experimental science which I hadn't experienced before and it was an instructive process to go through.

While waiting for the workshop to make the resonator, I simulated the results which we expect to get, using our model of how the SMM interacts with an applied electric field. These simulations indicate that with the microwave power available to us, the experiment should work, provided our model of the SMM is correct. In the last two days of my project, I was able to test the resonator at room temperature and found that it behaved almost exactly as expected. It was very satisfying to see the quite abstract theoretical simulations coming together with the very concrete constraints imposed by the size of the available components to produce a real object that actually worked.

Unfortunately, my project came to an end before all the parts for supporting the resonator inside the rest of the equipment were ready, so I was not able to test the resonator at low temperature or able to test the resonator with samples inside. However, I hope to go back in a few months when the experiment on the SMM will actually be carried out to watch and find out whether it works.

Through doing this project, I have gained experience of working as a more theoretically minded person in an experimental physics group. I have learnt to use a variety of scientific computing packages as well finding out what it's like to design real experimental equipment. I have also met several DPhil students and researchers working on quantum matter, which is a field in which I am interested in pursuing a PhD. Overall, this project has encouraged me to pursue a PhD after my undergraduate studies as I have enjoyed the experience of working in a research group.