Pulsed Electron Spin Resonance Studies of Atomic Clock Transitions in a Dimer of Cr$_7$Mn

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By Michael Cha

Advised by: Professor Charles Collett

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Amherst College
Department of Physics and Astronomy

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Abstract

Qubits, or quantum bits, rely on a quantum system that can hold any superposition of two states as opposed to just 0 or 1 as with a classical bit. Various systems have been explored as qubit candidates, including photons, trapped atoms, and both nuclear and electron spins. My work focuses on constructing two-qubit systems using dimers of molecular nanomagnets (MNMs), a class of magnetic material that can be chemically engineered to achieve various desired attributes. The focus of our current work, dimers of the MNM Cr$_7$Mn, features clock transitions between multiple spin states that increase the lifetime of the quantum state. We present pulsed electron-spin resonance (ESR) studies of dilute Cr$_7$Mn dimers in loop-gap resonators, including spectroscopic characterization of the dimer as well as progress on implementing two-tone ESR for two-qubit gates.
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Chapter 1

Introduction

1.1 Motivation

Quantum computers have recently become very popular, not only among physicists and computer scientists, but also among the general populace. Perhaps this is due to the proliferation of technology in general, and because it combines a common word, “computer,” with a widely known but generally less understood word, “quantum.” So what exactly is a quantum computer, and why is it so interesting to study?

A quantum computer, as its name implies, has essentially the same purpose as a computer, i.e. performing high-speed computations, but utilizes a different system of storing and processing information. In a classical computer, what we have in PCs, laptops, phones, etc., we make use of classical bits and logic gates. Bits store binary information as either 0 or 1, and logic gates process binary information by taking input bits and sending output bits according to some rule unique to the logic gate used. For example, an AND gate would produce a 1 if and only if both of two inputs are also 1. Any other combination of inputs would produce a 0. A NOT gate takes a single input and outputs the opposite, so a 0 would output 1 and vice versa. At the most basic level, this is how classical computers work.

Quantum computers make use of quantum bits, which we call qubits. Qubits are essen-
tially two-level quantum systems that we can place into one of the two levels or states, and use the state as the binary information. The difference here is that for quantum systems, the system can be in any superposition of the two available states. This is the main functional difference between a quantum computer and a classical computer, and what makes the quantum computer so desirable. Since the state can be in a superposition of states, it makes certain problems easier and faster to compute as opposed to a classical computer, such as creating and solving cryptographic systems.

Despite the nuance in storing information, quantum computers still make use of logic gates to process that information. It has been proven that CNOT (Controlled NOT) gates can be combined with single-qubit gates to have the same functionality as any other logic gates [1], so a first step in a quantum computer after acquiring viable qubits is to implement a CNOT gate, since doing so would mean that other types of gates and interactions would not have to be created individually.

The Friedman Lab’s and other groups’ previous research [2] on the viability of monomers of Molecular Nano-Magnets (MNM’s), as qubits. Their research has so far supported their viability as qubits. The work described in this thesis studies a dimer of Cr$_7$Mn to see whether it is a viable system of two qubits. To this end, there are two main goal for this research. The first is to check if the MNM’s retain qubit-like properties as a dimer, and the second is to study the possibility of using the coupling between the two MNM components of the dimer to connect the qubits in a single system.

In the next chapter, Background, I will discuss some information and previous research necessary to understand how we analyze the data. After that, in chapter 3, Spin Echo Experiments, I will present the data I obtained for a diluted sample of Cr$_7$Mn using spin echo experiments, which will be described in Background. Then I will discuss bimodal resonators and spin echo experiments done with the bimodal resonators, which are necessary to excite both monomer components of the dimer simultaneously or separately, in chapter 4, Bimodal Spin Echo Experiments. In chapter 5, I will present a summary of the data and
analyzed results I got from the spin echo experiments and bimodal resonators.
Chapter 2

Theoretical Background

2.1 Molecular Nano-Magnet Qubits

Molecular Nano-Magnets (MNM’s) are molecules that behave like magnets at zero field. They exhibit quantum behaviors at low temperatures, making them potential candidates for quantum bits, or qubits.

In the case of Cr$_7$Mn, there are seven chromiums and a manganese forming an octagonal ring as a base.

Each of the eight base atoms in Figure 2.1 (spin 3/2 for each Cr and spin 5/2 for the Mn) has a spin associated with it in one of the two possible spin states (positive or negative).

![Figure 2.1: A model of a Cr$_7$Mn MNM. The pink/purple sphere represents the Mn while the other 7 green spheres represent the 7 Cr’s.][2]
which we can represent here as up or down. By arranging the atoms up or down appropriately, the total giant spin (an approximation of the total spin of the molecule) results in a spin 1 \[2\]. The giant spin can then be treated as a single quantum spin \[2\]. Since they can be engineered to have certain atoms and arrangements, some of their properties, like the total spin, can be controlled. By using the giant spin as a single quantum system, we can represent the different states with an energy diagram. There are three energy states for this system, but it can be truncated to only two viable energy states, thus resulting in a two-state quantum system \[2\], which is necessary for a qubit to store binary information. Other requirements include a scalable physical system with well-characterized qubits, long relevant decoherence times, a “universal” set of quantum gates, and a qubit-specific measurement capability \[1\]. For the dimer system, since there are now two such spin 1 quantum systems, there are a total of nine states, or four in its truncated form. The Hamiltonian for a single monomer of Cr\(_7\)Mn is given by the equation:

\[
\mathcal{H} = -DS_z^2 + E(S_x^2 - S_y^2) - g\mu_B B \cdot S
\]  

(2.1)

For the dimer, as shown in Figure 2.2, there are now two of these monomers and a coupling between them. This gives the new Hamiltonian:

\[
\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + J_{parallel}S_{z1}S_{z2} + J_{perpendicular}(S_{x1}S_{x2} + S_{y1}S_{y2})
\]  

(2.2)

As seen in the left image in Figure 2.2, each monomer component has a different energy and thus different resonant frequency, as illustrated in the energy diagram on the right.

The decoherence time is essentially the length of time a qubit remains in a particular state. Thus, this is the amount of time available for a quantum computation to run a logic gate, since the gate cannot operate correctly if a state is not well-defined. In addition, we would like to be able to operate multiple gates sequentially, so longer decoherence times
become even more important.

The Friedman Lab recently did research on monomers of Cr$_7$Mn, a type of MNM, and verified that the molecules exhibit a sufficiently long decoherence time and an atomic clock transition at zero-field [2], which will be further discussed later.

### 2.2 Electron Spin Resonance (ESR)

To further understand decoherence times, we must first explain Electron Spin Resonance (ESR). For a powder sample of the dimer, we treat each molecule of Cr$_7$Mn as a single spin, so a collection of molecules would be represented as a collection of spins. With no external influence, given that the spins are not close enough together to affect each other, the spins should all be random, pointing in different directions in space, which we map on a Bloch sphere, a type of representation where a spin starts at the origin of the unit sphere and the orientation, or state of the spin is shown by an arrow from the origin. An example is shown in Figure 2.3.

A pulse is sent to the spins that rotates the spins 90 degrees away from the z-axis. Now that the spins are away from the z-axis, they will start to precess around the z-axis, but not all at the same rate due to slight differences in their spin Hamiltonians.

After some amount of time $\tau$, the spins are then flipped 180 degrees. This causes the
**Figure 2.3:** A Bloch Sphere. A model representation of a collection of spins and their orientations.

**Figure 2.4:** A model representation of a collection of spins and their orientations as they start to precess around the z-axis, or the direction of the external magnetic field.
precessing spins to precess back together, so even if they are precessing at different rates, they will realign for brief moment. When the spins realign, they produce a relatively large signal, called a spin echo [3]. The echoes are a result of refocusing precessing spins using a pulse sequence called a Hahn echo, and eventually the precessing spins will become random due to environmental factors, thus causing the echoes to decay. The amount of time it takes for the Hahn echo to decay is the decoherence time. Studies from the Friedman Lab have shown that the monomer of $\text{Cr}_7\text{Mn}$ has a decently large decoherence time of up to about $3.5 \mu s$, with an atomic clock transition [4], another important quality in a qubit.

### 2.3 Maximizing Signal and Rabi Oscillations

The echo signal becomes further reduced due to the filters, so it is essential that the echo signal size is as large as possible. Since phase is cyclic (0-360 degrees), there is a fixed range of values, and it is not apparent which phase works best for each sample until the echo signal is found, so the best phase is simply found through trial and error. By continuously applying Hahn echo pulse sequences and reading the echo signal as the phase is changed, we can determine what the best phase is for the sample. Once this is found, Rabi oscillations are used to determine the best lengths. Rabi oscillations plot the echo signal size as a function of a varying pulse length. The pulse length chosen can be either the first or second pulse of the Hahn echo sequence.

Once the signal has been maximized, we can finally take the data necessary to observe the decoherence times. Delay sweeps are taken to collect Hahn echo data from the sample over a discrete range of delay times, providing the data necessary to fit decay functions and calculate decoherence times. By plotting the area under the echo signals as a function of $2\tau$, the time taken between pulses, we can fit exponential decay functions with the decay times equal to the decoherence times. The spin echo data are taken at multiple magnetic fields near zero field, and we are looking to see if the atomic clock transition persists.
2.4 Qubit Systems and CNOT Gates

As mentioned in the last section, a quantum computer consists of qubits to store the information and logic gates to process the information.

For the logic gates, we want to create a CNOT gate, since it has the potential to build upon itself into any other type of logic gate [1]. Since quantum computing technology is still in its pioneering stages, it takes a lot of research and experimentation to create a system of interacting quantum systems that behaves in a particular way.

A CNOT gate is represented by the symbol given in Figure 2.5. Logic gate symbols are canonically read from left to right, so on the left there are two inputs and on the right two outputs. CNOT is short for Controlled NOT. This is because the bottom pair of input and output can behave either as a NOT gate or not a NOT gate (identity). When the upper input is 0, the bottom pair does nothing and outputs the same as the input. When the upper input is 1, the bottom pair acts as a NOT gate, and thus outputs the opposite signal of the input. The upper pair is purely to control the bottom pair, so it acts like an identity, outputting the same signal as the input.

By combining multiple CNOT gates in specific ways, other types of gates can be created. For example, by taking four CNOT gates and combining them as shown in Figure 2.6 where the green control inputs are always set to 1, an XOR gate is born. The red outputs are byproducts of the implementation, and the black inputs and outputs are the variables of interest. By following the logic of the gates, the output z is 1 when only x or y is 1 and the
Figure 2.6: A combination of 4 CNOT gates that form an XOR gate.

other zero. If x and y are both 0 or both 1, z is 0. Thus, we now have an XOR gate. In this way, every other type of gate can be created. While the same sequence of quantum CNOT gates would not create the same effect, a similar idea holds where single-qubit gates can be used with the quantum CNOT gates to perform other gate operations.

It is evident from the structure of the CNOT gate that two inputs, i.e. two interacting qubits are necessary. The dimer sample has two MNM’s, which we hope retain their long coherence times, and a coupling which theoretically serves as the interaction used to form the gate. To study if this is possible, the first step is to check if the dimer does indeed retain long coherence times, and then to be able to modify the states of each system separately with the same setup. To that end, we developed bimodal resonators which will be further discussed later.
Chapter 3

Spin Echo Experiments

We want to test the viability of the dimer sample as a system of qubits. For this we first need to verify that the components of the dimer retain long coherence times and a clock transition. To do this, we first performed some spin echo measurements on both MNM’s of the dimer. The measurements necessary are Rabi oscillations to find the largest signal size and Hahn echo experiments to find the echo signals, which will be further described here.

3.1 Experimental Setup

A powder of the dimer sample is first diluted in solution with toluene. This is done for two reasons. One is to make it easier to store and position the sample, since the solution can easily be trapped in an air-tight glass tube. In addition, the tubes can easily be replaced like cartridges, making any necessary changes to the sample very quick and easy. The other is to space the individual dimers away from each other. This reduces the effects of nearby dimers, and theoretically increases decoherence times as demonstrated by Kai Ellers in his research. The dilution is done volumetrically, so the percentage gives the spatial ratio of sample to toluene. Thus, the higher dilution, the greater the expected decoherence time. However, there is also the effect of decreasing signal size, since there will be fewer dimers in the sample. The sample solution is then positioned such that it is in the center of the loop
Figure 3.1: A simple diagram of a basic loop gap resonator on the right. On the left is a circuit diagram for an LC circuit, from which the resonator design is based on. These are usually made with Q-values around 100.

of a loop-gap resonator, as depicted on the right in Figure 3.2.

To check if the two MNM components of the dimer still behave as qubits, they must first be studied separately. To check for a large Hahn echo signal and decoherence time, as well as a noticeable atomic clock transition, there are a few steps that must be taken.

The first is to find the magnetic field at which the largest echo signal is found. The field was found via simulation. For our sample, the greatest echo signal was at a field of roughly 30 Oe, though we expect this to be due to a remnant field from the external magnet, and that the actual field is at 0. The resonant frequencies are at roughly 4.0 GHz and 5.3 GHz.

In addition to these factors, another parameter that affects signal size is the phase and pulse lengths. These must be checked experimentally.

A loop-gap resonator is essentially an LRC circuit where the resulting RF magnetic field is concentrated at the center of the loop. The loop is the inductor while the gap is the capacitor. Thus, by changing the properties of the loop and gap, the resulting resonant frequency of the circuit changes. For example, the gap thickness changes the dielectric thickness to tune the resulting frequency. These parameters are used to adjust the resonant frequency of the resonator such that it is close to the resonant frequencies of the MNM’s
in the dimer. With this setup, the pulse sequence, described in section 3.3 is sent via an antenna to the resonator, and if the sample is in the center of the loop with a similar resonant frequency, the sample resonates and sends back an echo signal. Since the echo signal is generated by the resonance, the signal we read is very small.

### 3.2 Rabi Oscillations

As explained earlier in section 2.2, the decoherence time of the quantum states of the dimer must be long enough to last the duration of the many logic gates. To better observe the decoherence time, we want to maximize the echo signal by choosing the correct experimental parameters. One of these parameters is the pulse length, which we find using Rabi oscillations as explained in Background.

We started with a 10\% solution at 1.9K used Hahn echo sequences to observe the echoes. We then ran a Rabi oscillation measurement, and obtained the data given in Figure 3.3. For this measurement, we fixed the first pulse length and varied the backpulse length. We use the x-position (length of pulse) of the highest point of the plot as our pulse length, since that gave us the maximum echo signal. This value was found for this part of the dimer to be approximately 120 nanoseconds.

![Figure 3.2: An LGR with the sample.](image)
Figure 3.3: The Rabi Oscillations analyzed from the 0.001% solution on the 4.0 GHz MNM with a 4.313 GHz resonator, 1.9K and zero field. Signal size as a function of the pulse length. Each curve represents an echo taken at a different delay time, with the earliest delay times having the largest signal and gradually dying down.
Figure 3.4: The echo signals from the 10% solution for the 4.0GHz component at 1.9K and a range of $\tau$.

### 3.3 Hahn-Echo Signals

The resonant frequencies that we expect for the dimer samples were roughly 3.9 GHz and 5.29 GHz. Since the 4.0 GHz LGR resonance was similar to the resonance of the monomer, we started the Hahn echo measurements with this resonance, hoping to compare with the results for the monomer samples.

We ran delay sweeps as described in the Background at multiple external magnetic fields to get a set of decoherence times. Figure 3.4 shows the raw echo data for these measurements. To find the signal size, we used the area under the signal, by first aligning each echo so the signals were centered at the same point, and then choosing a window that included the start and end of the signals over which to integrate to find the area.
Figure 3.5: Echo size vs the delay times are fit to exponential decay functions for each set of data from various magnetic fields. This data is from the 10% solution 4.0 GHz component at 1.9K.
Figure 3.6: The decoherence times ($T_2$) with respect to the external magnetic field. This data is from the 10% solution with the 4.3 GHz resonator at 1.9K. The error bars show the deviation of the data from the decay fits, probably due to slight temperature fluctuations, difference in resonant frequencies between the resonator and sample, and various other environmental effects.

To analyze these data, we plotted the area under the echo against $2\tau$, or twice the delay time, in order to show the decay of the time evolution of the signals. Plotting this data shows the decay of the echoes taken at different magnetic fields, as shown in Figure 3.5. These decays are then fit to exponential decay functions. The time constants derived from these exponential decay fits are the decoherence times, which we plot with respect to the magnetic fields in Figure 3.6. There is a clear clock transition here identified by the unimodal peak as was also seen in the monomer data, which show that these features persist for the lower frequency component of the dimer.

The clock transition is seen at non-zero field, in this case at around 30Oe. We believe
this to be due to a remnant field in the external magnet, and that the clock transition is actually at zero field.
Figure 3.7: The decoherence times ($T_2$) with respect to the external magnetic field. This data is from the 0.001% solution with the 4.3 GHz resonator at 1.9K.

We then ran these experiments again with a higher dilution (0.001%) in an attempt to increase the decoherence time. From these we get the decoherence times in Figure 3.7 with a clock transition at 25 Oe (-5 Oe without the remnant field) and a maximum decoherence time of roughly 3.1 $\mu$s, which is noticeably larger than the maximum of about 2.1 $\mu$s for the 10% solution.

These measurements were then repeated for the higher frequency component, to check if the 5.3 GHz MNM component of the dimer also exhibits qubit-like properties. The resulting decoherence times are shown in Figure 3.8. These results still show a clock transition, but the peak is noticeable less sharp. This implies that the echo data collected here may not all be from the higher frequency component, and may suggest that the resonator is also resonating partially with some of the lower frequency population. Since the resonators have some range of frequencies that may drive transitions in the dimer, it is possible that both dimer components may fall within that range. The sizes of the error bars are also much
Figure 3.8: The decoherence times ($T_2$) with respect to the external magnetic field for the 5.0 GHz resonator. This data is from the 0.001% solution at 1.9K larger than that of the lower frequency component at the same dilution, further supporting the suspicion that the data may not be completely from the higher frequency component.
Chapter 4

Bimodal Spin Echo Experiments

In the last chapter, I discussed experiments done individually on the lower and higher frequency components of the dimer samples, which served to verify that the MNM components retained long decoherence times and an atomic clock transitions. Next, we need a system where we can simultaneously manipulate the states of either MNM in the dimer. For this, we need bimodal resonators.

4.1 Bimodal Resonators

The idea of a bimodal resonator is to create a single resonator “unit” of some sort that resonates at 2 different frequencies. This allows us to excite either component of the dimer without having to change the setup each time, which would not be practical for quantum computing.

The first design idea we tried was a single solid resonator with 2 sets of loops and gaps, like shown in the CAD drawing in Figure 4.1. Since a loop-gap resonator is basically just an LC circuit where the magnetic field gets concentrated in the inductor, this idea was designed to place 2 different LC circuits (see section 2.1) with different resonant frequencies in parallel, with the inductors spatially intersecting to create a region where the RF magnetic fields overlap. In this region both components of the dimer sample can be excited. As
explained in section 3.1, the parameters of the loop and gap affect the resonant frequency of the magnetic field. Thus, by having each set of loop and gap with different parameters (gap width, loop radius, etc.), there are two different resulting fields and frequencies.

The first prototype we created had a clipped design, like shown in the image in Figure 4.2. In this figure, the light blue cylinder is the model for the shield cavity in which the resonator will be placed. The dark blue body is the resonator. The simulated RF magnetic field is indicated by color in this 3D plot. While it is difficult to see this with this image, the field is present in the intersection between the loops. The two nested white and yellow cylinders are the antennae, though it was determined that only one antenna was ultimately necessary for this design.

To check the capabilities of this resonator design, it was placed in a shield with a single antenna, and when a signal was passed through the antenna, the resonant frequency was as shown in Figure 4.3. The presence of two prominent peaks, or resonant frequencies, validates the potential of this design for future experiments with the dimer system.
Figure 4.2: 3D design of clipped single-unit bimodal resonator with simulated magnetic field. This simulation used the right antenna as a port and used a frequency at around 4.7GHz.
Figure 4.3: The resonant frequencies of the bimodal resonator featuring two large peaks, or resonances. The frequencies are at roughly 4.75 GHz and 6.3 GHz.
4.2 Stacked Resonators

The other design idea was to use preexisting known loop-gap resonators and stacking them on top of each other. The concept behind this design is for an antenna to resonate both resonators together. To avoid coupling via electrical connection between the 2 resonators, we placed a small spacing between the resonators as shown in Figure 4.4.

We want the spacing to be large enough to avoid coupling between the resonators, but also small enough such that the magnetic fields that should be present in the center of the loops would also overlap in some small region between the 2 loops. Similar to what was seen in the magnetic field simulation for the bimodal resonator in Figure 4.3, the magnetic field tends to extend slightly outside of the cross-section of the loop. We use this to our advantage by overlapping the extended regions. This would essentially grant us the same bimodal capabilities as the single solid bimodal resonator design described earlier, but would also have the benefits of being easy to make with no need to design/build an entirely new resonator, and producing coaxial magnetic fields, allowing for easy alignment and placement of the sample. Also, because of the geometric limits of the region of con-
centrated magnetic fields, it is highly unlikely that more than two resonators can be stacked and produce a single region where all fields are concentrated. However, for our purposes, and for the purposes of potentially creating a CNOT gate, two frequencies are enough since this is a dimer sample with only two frequencies.

After testing the resonators without a sample to find the resonant frequencies, we saw 2 clear and distinct resonances shown in Figure 4.5. The frequencies of each resonator when tested individually were 4.3 GHz and 6.0 GHz, and the stacked resonator setup showed 5.02 GHz and 6.25 GHz. Both the frequencies appeared to shift, indicating some amount of coupling, since both frequencies were affected.

One requirement we discovered, however, is that the resonator placed on top, closer to the antenna, must be smaller than the bottom resonator, or clipped in some way. The reason for this is that when we tested with 2 completely circular resonators of the same radii, the
bottom resonator was being blocked out. This was due to the same concept behind the shield. The conductive surface of the upper resonator acted as a shield to block the signals coming from the antenna from reaching the bottom resonator. When we tested with a fully circular resonator on the bottom and a clipped resonator on the top, as we did in the setup in Figure 4.4, we saw the resulting 2 resonances in Figure 4.5.

### 4.3 Testing the Stacked Resonators

As a test for the stacked resonators setup, we first ran the Hahn-Echo experiments from the last chapter using the 0.001 percent solution again. Since the effective region of magnetic field is formed by the overlapping parts of each resonator’s magnetic field, the said region is naturally smaller than a region from a single resonator. This means that there will spatially be fewer dimer molecules in the effective region compared to with a single resonator, and thus the signal is expected to be smaller. Due to this and the observation that the resonant frequencies were slightly shifted compared to the individual resonators, we expect something slightly different but very similar to what was seen in the last chapter. At the very least, we expect the atomic clock transition to persist and to be at roughly zero-field.

From these experiments using the 5.02 GHz component of the resonator we get the data in Figure 4.6. We attempted to select for the 5.02 GHz component by applying filters. From these we get the decoherence times in Figure 4.7, and see that the atomic clock transition still exists at 25Oe, slightly off from the theoretical zero-field, but expected because of the remnant field from the external magnet. It should also be noted that the error bars are larger for this measurement than for the single component resonator. This is likely due to the shift in resonant frequencies of the resonances from the resonant frequencies of the sample, which would result in a broader range of decoherence times since the resonances of the resonators are further from the resonant frequencies of the dimer. In addition, the
Figure 4.6: Decay data and fits from the lower resonance of the stacked resonator setup.

Echo signal sizes were found to be smaller than for the experiments with the regular LGR’s, thus decreasing the signal-to-noise ratio and increasing the error.

When checking for the best phase to use with the 6.25GHz component of the resonator, we managed to find a strong signal at the same phase of about 115 degrees twice, but after that we could not replicate it. We believe there might be a mixture of external issues with these experiments. One possible issue is with our PPMS (Physical Property Measurement System), which we use to control the temperature of the sample. Recently while these experiments were taking place, the temperature has been somewhat unstable and harder to control, thus resulting in higher temperatures for the sample at random times while we took the data. We believe this to be a likely potential problem with these experiments.
Figure 4.7: The decoherence times from the lower resonance of the stacked resonator setup.

4.4 Future Steps

Now that we have working bimodal resonators and have verified that both MNM components of the dimer have clock transitions and sufficiently long decay times, the next step is to use the bimodal setups to control one of the MNM states by manipulating the other. If we succeed in controlling the state of one MNM based on the state of the other MNM, we will effectively have a base from which to build a CNOT gate.
Chapter 5

Conclusion

There were two main goals for this research: check if the monomer components to the dimers kept their long coherence times at a clock transition and study whether we might be able to use the coupling between the two monomer parts to form a qubit system. There are still experiments and tests to run to satisfy the second goal, but promising data has been collected with regards to the first.

The dimer in question is based on two different monomers of Cr$_7$Mn, and the lower of the two resonant frequencies of the dimer is the same as the monomer studied by Collett et al [2]. Because of these similarities, we first checked the lower frequency component for qubit viability. At a volumetric dilution of 100,000, or a 0.001\% solution, the lower frequency component showed a decoherence time of up to around 3.0 microseconds with a clear clock transition at effectively zero-field. This long decoherence time and the presence of the clock transition are consistent with the results found from the monomers.

The higher frequency component was also tested. Due likely to a combination of potential issues in our setup, including the PPMS, the filters, and possibly even the dilution of the sample, we were only able to find and replicate a decent echo signal once, after which only noise was found again. Thus, since the data seemed corrupted, we cannot conclude anything about the higher frequency component at this time.
For the second goal, while all the necessary tests have not been run yet, promising methods and setups have been found for implementing the future experiments towards this goal. Two potential bimodal resonator systems have been identified. To test the viability of a dimer as a qubit system and not a clump of individual and disparate qubits, both components of the dimer sample must be excitable within the same experiment. They need to both be controllable individually and simultaneously, which can be enabled by using a bimodal resonator. The first of these systems is a single resonator unit that has two sets of loops and gaps. Simulations and a characterization test have produced multiple distinct frequencies for this resonator.

The second design is the stacked resonator, which is made by taking multiple 2D loop-gap resonators and literally stacking them on top of each other. This setup resulted in two distinct and clear resonances. This is the system that was used for the bimodal spin echo experiments. While not all experiments have been run with this setup due to the technical difficulties mentioned earlier, it did prove to work as expected for the 5.02GHz component of the resonator, and thus makes it a viable bimodal resonator system for future use.

Future experiments for this research involve probing the coupling within the dimers. Next is implementing a CNOT gate using the bimodal resonator system. This can be accomplished if we are able to control the state of one of the MNM components of the dimer by manipulating the state of the other MNM. Should this be accomplished, the next steps would be to combine the CNOT gates into other logic gates and ultimately build a quantum computer.
Appendix A

Data Acquisition Protocols

A.1 Simulations

To characterize the dimer samples, we first simulated experiments on the sample to get an idea of where to look for resonances. For our purposes, we used EasySpin, a quantum physics toolkit on MATLAB. By inputting the necessary known values of the spin hamiltonian for the dimer and proper experimental conditions, the simulation will tell us where we can expect to find the resonance and roughly at what field that signal is the strongest.

A.2 Setup

For the basic components of our setup, we start with a signal generator which sends the pulses. These pulses go through a circulator, which acts as a one-way street (1 outputs to 2, 2 outputs to 3, and 3 outputs to 1, but not vice versa). The pulses comes out at 2 and enters the probe that contains the sample. The pulses resonate with the loop-gap resonator within the conductive shield protecting the resonator and sample via the antenna, and the resonator emits a magnetic field in the loop where the sample is held (see Chapter 3 for more details), which then resonates with the sample and given that the frequency of the sample and the frequency of the resonator match, the resulting echo signal is then sent back through the
antenna into port 2 of the circulator, which is then outputted to port 3 of the circulator. This signal is then read from an oscilloscope and analyzed.

### A.3 Sample Preparation

To do these measurements, we first have to prepare the samples. The dimer samples are in powder form. The powder is then volumetrically diluted with toluene to space individual dimers from each other. The solution is then loaded into a small glass tube which can be fitted into the loop of the loop-gap resonator as described in section 3.1. The sample is then fixed to the shield that contains the resonator such that the solution is in the center of the resonator’s loop, after which the experiments can be run. The antenna is attached to the cap of the shield such that it is close to but does not touch the resonator or sample. The shield, antenna, resonator, and sample setup is then attached to a probe, which is lowered into a cryostat and cooled to low temperatures to reduce the effects of temperature and emphasize quantum effects. The spectrometer connects to the probe so that input signals can be sent into the shield and reflected echo signals can be read from the spectrometer.

### A.4 Data Correction

Hahn echo data was taken every time we wanted to find the decoherence times. Unfortunately, there is a lot of noise relative to the signal that might affect data analysis. In case there might have been an offset in the data, we also subtracted every signal by the average of the last 50 points of data from the echoes. The last 50 points, as can be seen on the right end of the signals in Figure A.1, are not flat due to noise, but since we expect noise to be random, can be averaged to roughly what we expect for the offset. Since the signal is still small, with a decent amount of noise, the area was chosen for the signal size as opposed to the peak of the signal to further reduce the effect of noise.
Figure A.1: The echo signals from the 10% solution for the 4.0 GHz resonator at 1.9K and a range of fields near 0.

