Magnetic Spin Correlations in $NaYbO_2$ at Near-zero Temperatures

Kristina Nuttall

A senior thesis submitted to the faculty of
Brigham Young University
in partial fulfillment of the requirements for the degree of

Bachelor of Science

Benjamin Frandsen, Advisor

Department of Physics and Astronomy

Brigham Young University

ABSTRACT

Magnetic Spin Correlations in $NaYbO_2$ at Near-zero Temperatures

Kristina Nuttall
Department of Physics and Astronomy, BYU
Bachelor of Science

Sodium Ytterbium Oxide ($NaYbO_2$) is a geometrically frustrated magnet that has generated interest as a potential quantum spin liquid and could have applications in quantum computing. We are interested in its magnetic structure at temperatures near zero Kelvin, where the correlations unique to a quantum spin liquid would be most prominent. Through analysis of neutron scattering data for $NaYbO_2$, we determined that it exhibits antiferromagnetic correlations between near-neighbor spins at 3 K, which decrease at higher temperature but remain nonzero up to 25 K. The nearest-neighbor correlations dominate the neutron scattering signal, but contributions from further neighbors are also present. Additionally, the neutron scattering data can be fitted well to both a two-dimensional and a three-dimensional magnetic model. These results are consistent with the expectations for a quantum spin liquid, and although we cannot prove definitively that $NaYbO_2$ is a genuine quantum spin liquid, this work provides important new insights into the magnetism of this interesting compound.

Keywords: geometrically frustrated magnet, quantum spin liquid

ACKNOWLEDGMENTS

I would like to thank my undergraduate research advisor, Dr. Frandsen, for his encouragement and assistance in my research process and in my writing this paper. I would also like to thank my husband Jacob Nuttall and my family for their loving support.

Contents

Chapter 1

Introduction

In this section we discuss the meaning of geometrically frustrated magnetism and the distinctive properties of quantum spin liquids, as well as the structure of sodium ytterbium oxide ($NaYbO_2$). We then give a brief summary of the experiment and results.

1.1 Geometrically Frustrated Magnets and Quantum Spin Liquids

Magnetic materials are made up of many small magnetic moments comprised of the unpaired electrons in the atoms. In crystalline magnetic materials that are electrically insulating, these moments are located at periodic positions corresponding to magnetic atoms in the crystal structure. Since the magnetic moments for the material I will be discussing in this paper are produced by electron spins, I will be referring to them as spins. When two spins point in the same direction, we say that they are ferromagnetically aligned, and when they point in opposite directions, they are antiferromagnetically aligned.

Geometric frustration occurs when the structure of a magnetic material makes it impossible

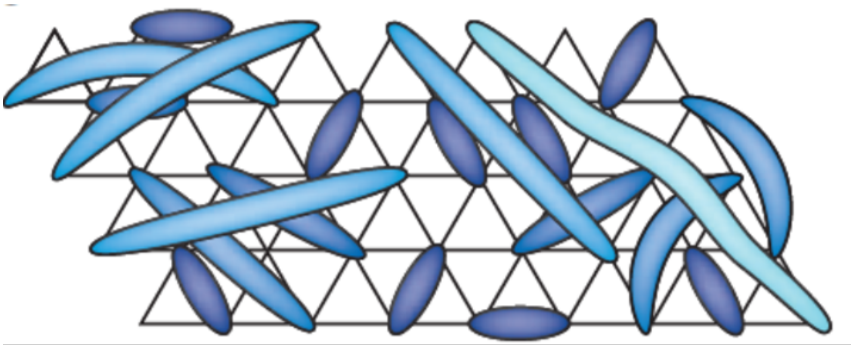

Figure 1.1 Schematic illustration of a quantum spin liquid state consisting of entangled correlations between spins on a triangular lattice. The true ground state of a quantum spin liquid would consist of a quantum superposition of many such configurations. Figure adapted from Ref. [1].

for all magnetic interactions between spins to be satisfied simultaneously. An example of this is a triangular lattice structure with antiferromagnetic nearest-neighbor interactions, since nearest neighbor spins are grouped in threes [1]. If one spin is up and another is down, then what should the third be? Either of its options, up or down, would create an imbalance in the total spin of the neighbors, so it does its best by fluctuating between the two; its neighbors do likewise. This leads to some unusual magnetic behavior. Whereas most magnetic materials' spins freeze into long-range order below some characteristic temperature, with each of its spins being correlated with other spins up to many cell spacings away, geometrically frustrated magnets can sometimes display quantum spin liquid behavior, where the spins dynamically fluctuate between degenerate configurations that are characterized by short-range correlations–those between a spin and just its first few nearest neighbors–even down to zero Kelvin. Furthermore, the spins have a high degree of quantum entanglement, despite not ordering into any long-range pattern (Fig. 1.1). Quantum spin liquids have potential applications in quantum computing because of this entanglement and the possibility of storing information in their spins.

1.2 Sodium Ytterbium Oxide, $NaYbO_2$

Sodium Ytterbium Oxide, $NaYbO_2$, was recently discovered and shown to be a geometrically frustrated magnet and thus a potential quantum spin liquid [2] [3]. It features a triangular lattice structure, with spins located at each ytterbium atom. Its lattice parameters—the dimensions of its crystalline cells—are 3.34556, 3.34556, and 16.45590 Angstroms, with angles between them of 90, 90, and 120 degrees [Figure]. We aim to learn more about the magnetic behavior of $NaYbO_2$ at low temperatures and how consistent it is with the quantum spin liquid model.

1.3 Experiment Summary

We performed low-temperature neutron diffraction experiments on a powder sample of $NaYbO_2$ to obtain magnetic scattering data. We then performed reverse Monte Carlo refinements on the data using Spinvert and Spincorrel and conducted additional modeling using magnetic pair distribution function (mPDF) methods. We determined that its magnetic structure can be modeled well using either a 2D or a 3D model, and it exhibits an antiferromagnetic correlation between first-nearest neighbors and likely some other significant correlations between further neighbors. These results are consistent with the expectations for a quantum spin liquid. However, they are not sufficient to prove that $NaYbO_2$ is one definitively.

Chapter 2

Methods

In this section we discuss the methods and programs used to obtain our experimental data and perform the analysis. First, neutron scattering, which is the experimental process that produces the magnetic diffuse scattering we analyzed. Second, Spinvert and Spincorrel, which are programs for modeling the diffuse magnetic scattering data using reverse Monte Carlo methods. Third, mPDF, which is a Python package for modeling the magnetic relations for a set of spins specified by the user.

2.1 $NaYbO_2$ Sample Synthesis

The sample of sodium ytterbium oxide ($NaYbO_2$) analyzed in this project was produced by our collaborators at Santa Barbara via a solid-state reaction according to the methods described in Ref. [4].

2.2 Neutron Scattering

Neutron scattering is a method for revealing the crystallographic or magnetic structure of a material. The material is placed in a beam of neutrons and the distribution of scattered neutrons is measured, forming a pattern dependent on the material's structure. The main experimental quantity of interest is the intensity of scattered neutrons as a function of scattering angle, which is related to the change in momentum experienced by the scattered neutron. Our neutron diffraction data was collected from a powdered crystal sample by our collaborator Henry Fischer on beamline D4 at the Institut Laue Langevin in Grenoble, France. He collected scattering data at temperatures between $2.4K$ and $50K$. The data at $50K$ contained no contributions from coherent magnetic correlations, since the thermal energy at that temperature is sufficient to disrupt any spin-spin correlations. Therefore, the data collected at $50K$ consisted only of temperature-independent scattering from the nuclei. This scattering pattern was subtracted from the data collected at lower temperatures to remove the nuclear scattering, leaving only the diffuse magnetic scattering as the difference.

The neutron scattering experiment was performed by our collaborator Henry Fischer at the Institut Laue Langevin in Grenoble, France, at temperatures between 3 K and 50 K. The data at 50 K contained no contributions from coherent magnetic correlations, since the thermal energy at that temperature is sufficient to disrupt any spin-spin correlations. Therefore, the data collected at 50 K consisted only of temperature-independent scattering from the nuclei. This scattering pattern was subtracted from the data collected at lower temperatures to remove the nuclear scattering, leaving only the diffuse magnetic scattering as the difference.

2.3 Spinvert and Spincorrel

Spinvert and Spincorrel are computer programs for analysis of magnetic materials [5]. Spinvert takes in a set of experimental magnetic diffuse scattering data along with a configuration document,

which is produced by the user and details the structure of the magnetic material for which the data was taken. It then employs a reverse Monte Carlo approach to come up with a hypothetical array of magnetic spins that would produce a scattering pattern similar to the experimental data it was given. The reverse Monte Carlo approach is an iterative process where the program tries altering a spin, checks the new fit against the data, and keeps the change if it improved the fit. The program even accepts with a certain probability spin moves that make the fit worse, which allows it to escape local minima in the fitting space. This is done in turn for each spin repeatedly until the fit is a good enough approximation of the data.

Spincorrel takes in the spin file produced by Spinvert and calculates the spin correlation function via the equation below (Eq. 2.1).

$$\langle \mathbf{S}(0) \cdot \mathbf{S}(r) \rangle = \frac{1}{N} \sum_{i=1}^{N} \sum_{j=1}^{Z_i(r)} \frac{\mathbf{S}_i \cdot \mathbf{S}_j}{\langle Z_i(r) \rangle} \tag{2.1}$$

The spin correlation function tells us how strongly the nearest-neighbor, next-nearest-neighbor, etc. spins are correlated, and whether that correlation is ferromagnetic or antiferromagnetic.

2.4 mPDF

The magnetic pair distribution function (mPDF) is another approach to determining short-range magnetic correlations from diffuse magnetic scattering. Typically, this method involves Fourier transforming the scattering data to analyze the local magnetic correlations in real space. In this paper, however, we took a slightly different approach by calculating the real-space mPDF for a given spin configuration and then Fourier transforming back into reciprocal space to compare to the data. This allowed us the flexibility of defining custom magnetic models in real space but making the comparison to our data in reciprocal space. We used the python package diffp.mpdf to do this [6].

Chapter 3

Results

In this section we discuss what was observed about both the overall magnetic structure of $NaYbO_2$ as well as the short-range correlations between neighboring spins.

3.1 2D Versus 3D Model

The crystal structure of $NaYbO_2$ can be described as sheets of magnetic spins in a triangular lattice. One of the first questions we had regarding the magnetic behavior of $NaYbO_2$ is how significant the interactions of spins between its layers is. This is relevant because the QSL state in $NaYbO_2$ is expected to contain only magnetic correlations within the two-dimensional plane of each layer. To analyze this, we ran two separate instances of the program Spinvert, using the same data for both but different configuration files for each. One configuration file listed the real lattice parameters for $NaYbO_2$. The other featured an alteration to the parameter for the distance between sheets, increasing it by a factor of about 10. This change forced the program to disregard the possibility of inter-planar spin interactions, constraining it to what we call a 2D model. The unaltered parameters describe a 3D model.

We plotted the fits with the scattering data to get a qualitative measure of the goodness of the

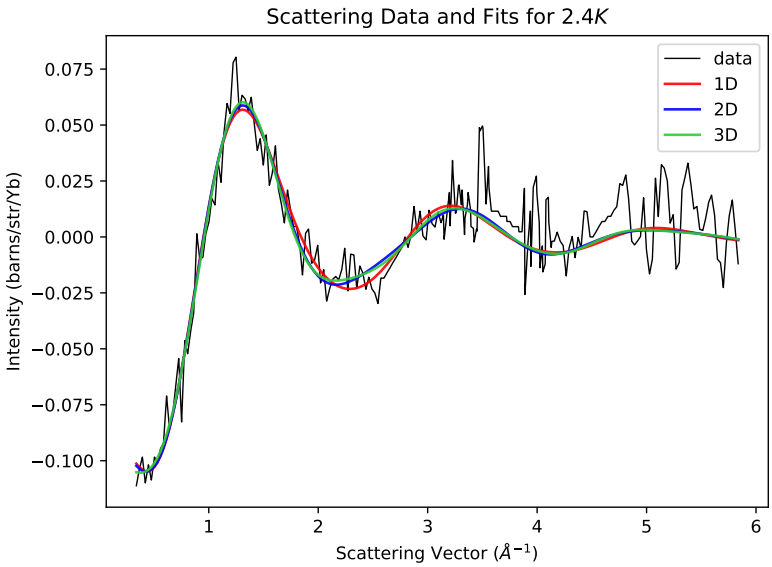

Figure 3.1 Fits produced by Spinvert to the neutron scattering data for $NaYbO_2$ at $2.4K$ for 1D, 2D, and 3D spin structure models. The three fits are slightly different, but each matches the data quite closely.

fits. As can be seen in seen in Fig. 3.1, both the 2D and 3D fits closely match the data, which leads us to conclude that the magnetic structure of $NaYbO_2$ can be modeled as either 2D or 3D equally well. Thus, the data is consistent with the 2D model expected for the QSL state. We also tried a third configuration with two of the three axes enlarged to represent a 1D model, and it also seemed to match the data well, which was surprising.

In order to get a quantitative idea of the goodness of the fits, we calculated the R values for each fit (Eq. 3.1).

$$R = \sqrt{\frac{\sum_Q \left[(I_{calc}(Q) - I_{expt}(Q))/\sigma(Q)\right]^2}{\sum_Q \left[I_{expt}(Q)/\sigma(Q)\right]^2}} \tag{3.1}$$

To more easily compare the R values, we calculated the fractional difference between the 1D and 2D values and the 3D values for each temperature (Fig. 3.2). Although the R values for the 3D model are consistently the smallest out of the three models, and the ones for 1D are consistently the largest, the 1D and 2D values are different from the 3D ones by less than 4% at all temperatures.

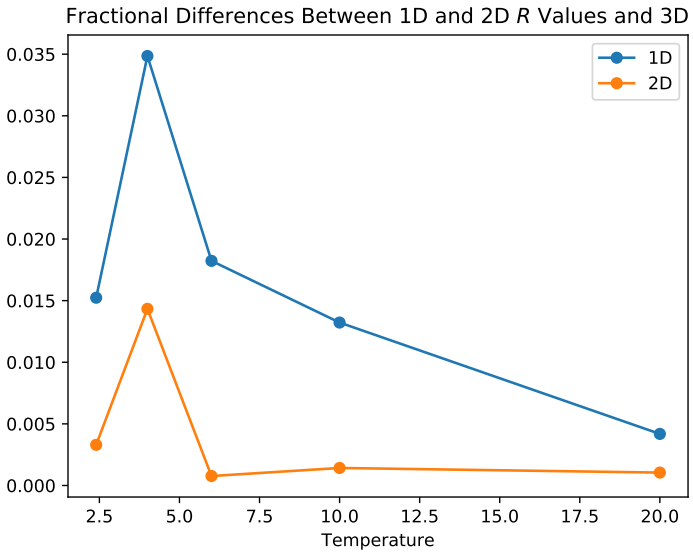

Figure 3.2 Fractional differences between the R values of the 1D and 2D fits and the 3D fits for each temperature. The R values of the fits are very close, never more than 4% different, indicating that the 1D, 2D, and 3D models all produce similarly good fits to the neutron scattering data.

The 1D fit being a good match for the data was unexpected since $NaYbO_2$ does not have a favored geometric direction because it is geometrically frustrated. This result is instructive, as it demonstrates that there are multiple spin configurations that can produce good fits to our neutron scattering data, even ones whose structure is radically different from that of $NaYbO_2$. This demonstrates that caution must be used when interpreting the fitting results to avoid making overly strong conclusions, and that physical knowledge of the structure of the material must be used to select reasonable models. In this case, the 1D model can be disregarded as non-physical, even though it provides a fit nearly as good as the 2D and 3D models.

3.2 Spin Correlations

Our next question was whether the spins in NaYbO2 tend to be ferromagnetically or antiferromagnetically aligned with their nearest neighbor spins, and how strong those correlations are at different temperatures. To answer this question, we used Spincorrel to calculate the spin correlation function (scf) for each of the 3D spin configurations generated by Spinvert (Fig. 3.3). The scf gives the average correlation between a single spin and each of its first few nearest neighbors, positive for ferromagnetic and negative for antiferromagnetic. These correlations are given as a function of the distance between the spin and the neighbors. We found that $NaYbO_2$ exhibits an antiferromagnetic first-nearest neighbor correlation that is stronger at lower temperatures (Fig. 3.4). This makes sense because increased thermal energy introduces random variations that make the interactions less pronounced. Beyond the first-nearest neighbor the correlations are uncertain: while the scf gives values for those correlations, their associated errors are so big that they could feasibly be either antiferromagnetic or ferromagnetic.

3.3 Agreement to a Single Pair of Spins

This brings us to our final question, which is if there are any significant interactions between farther neighbors or if the first-nearest neighbor correlation is sufficient to represent the magnetic structure. To answer this, we checked the agreement between the scattering pattern of $NaYbO_2$ and a single pair of antiferromagnetically aligned spins, which we produced using mPDF (see the orange curve in Fig. 3.5). The agreement is surprisingly good considering the simplicity of the model, which helps justify the 1D model's fit quality. However, it seems to be missing some details in the function shape, leading us to conclude that there exist significant correlations between neighboring spins beyond the first, but the RMC analysis cannot unambiguously determine what they are.

The fact that non-trivial spin correlations exist beyond the nearest neighbor is important for

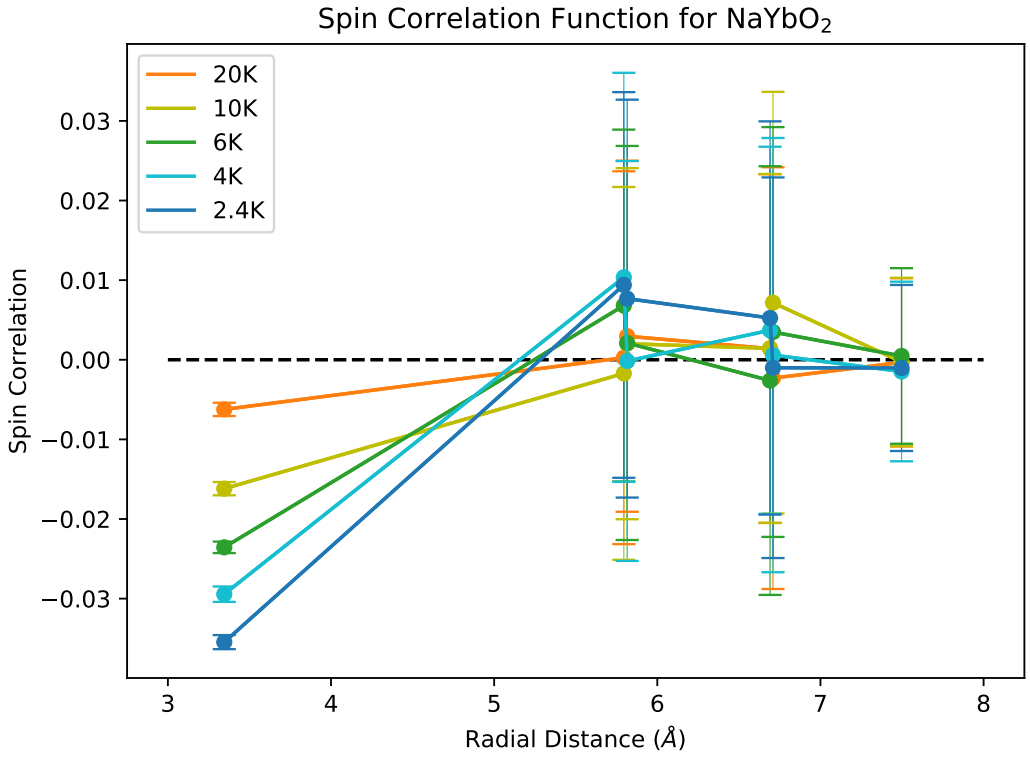

Figure 3.3 Spin correlation functions produced by Spincorrel based off the spin configurations produced by Spinvert for $NaYbO_2$ using a 3D model. The horizontal axis represents the distance between a spin and its nearest neighbor spins. The vertical axis represents the average correlation between a spin and its neighbor, positive for ferromagnetic and negative for antiferromagnetic. The first-nearest neighbor correlation is well-defined, but further correlations have too much error to be determined.

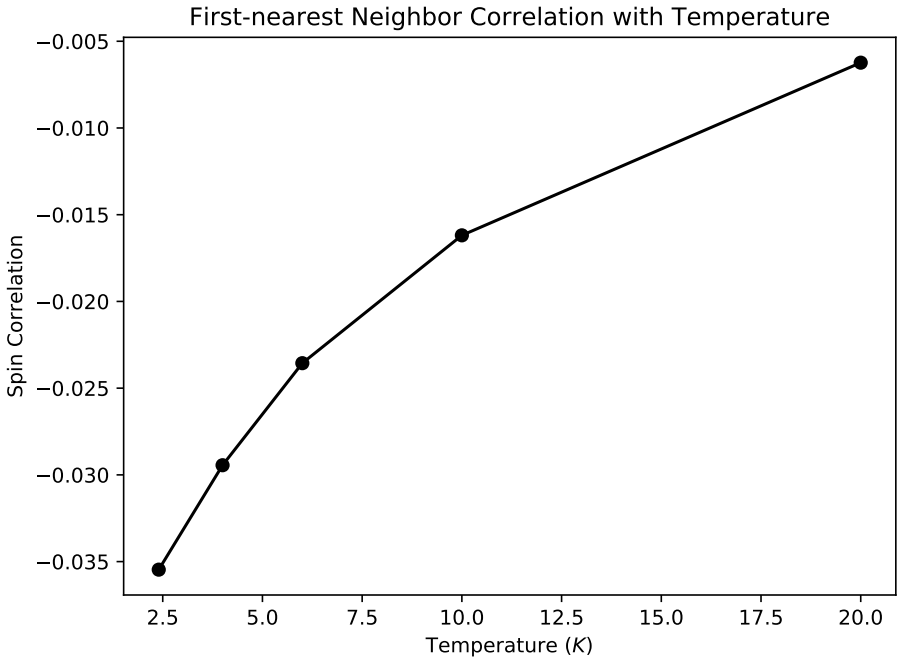

Figure 3.4 The first-nearest neighbor spin correlations from Fig. 3.3 plotted against temperature. The correlation is antiferromagnetic and stronger at lower temperatures.

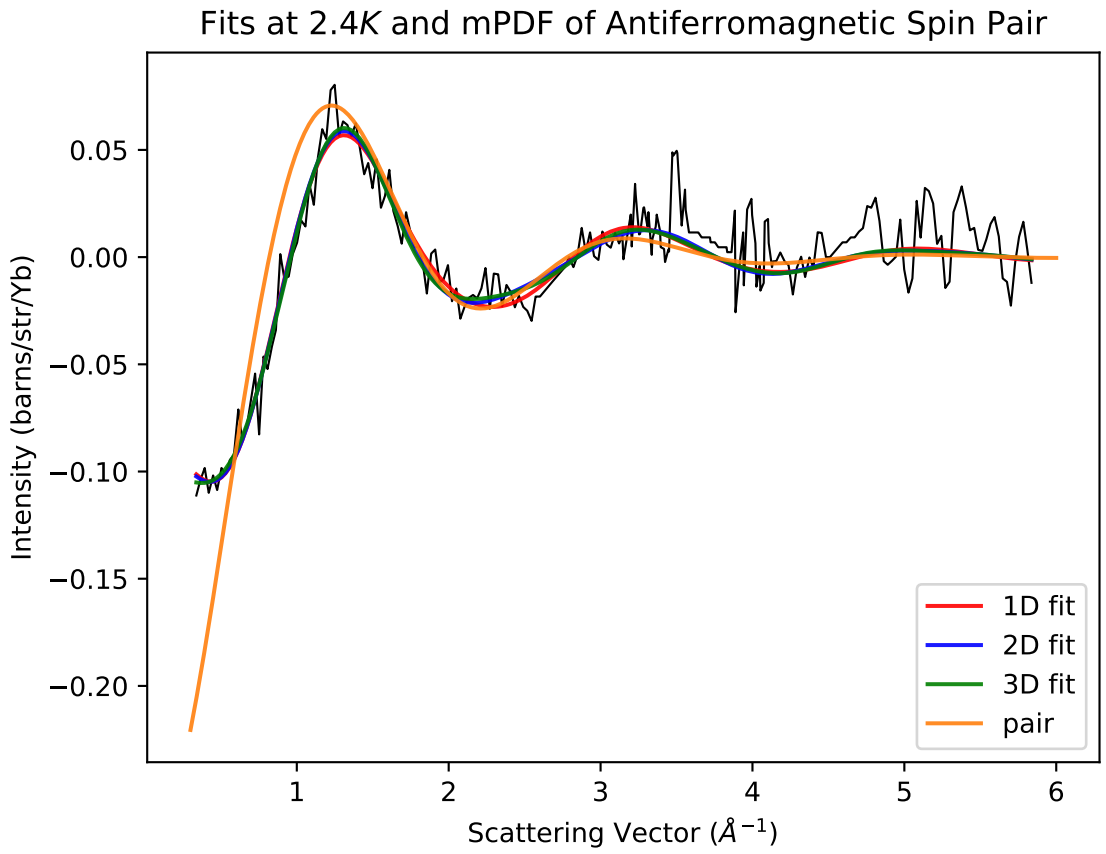

Figure 3.5 The simulated neutron scattering pattern for a single pair of antiferromagnetically aligned spins produced using mPDF along with the scattering fits and data for $NaYbO_2$ at $2.4K$. The scattering for the pair of spins matches the general shape of the data, but is missing some of the finer details. This leads us to conclude that there exist correlations between spins farther apart than first-nearest neighbors that are significant in causing the observed scattering pattern.

the discussion of NaYbO2 as a potential QSL material. The QSL ground state contains complex correlations caused by the entanglement of spin pairs over a range of distances extending beyond just the nearest neighbor spins. As such, the diffuse scattering pattern corresponding to such a state should be more complicated than simple nearest-neighbor antiferromagnetic correlations. We have demonstrated here that this is indeed the case, since the simulations using a single pair of spins is unable to capture the details of the shapes and positions of the peaks in the diffuse scattering pattern.

3.4 Conclusion

The neutron scattering data and reverse Monte Carlo analysis presented here provide a deeper understanding of the magnetic correlations present in the quantum spin liquid candidate material $NaYbO_2$. The results show the presence of short-range magnetic correlations dominated by nearest-neighbor antiferromagnetic interactions, but with nonzero correlations for more distant neighbors as well. These observations are all consistent with what would be expected for a quantum spin liquidf ground state. However, the data do not permit an unambiguous determination of the magnetic correlations beyond the nearest neighbor, nor do they constrain the dimensionality of the correlations. For this reason, further work is required to determine whether $NaYbO_2$ could truly have a quantum spin liquid ground state.

Bibliography

[1] L. Balents, "Spin liquids in frustrated magnets," Nature **464,** 199–208 (2010).

[2] W. L. et al, "Rare-Earth Chalcogenides: A Large Family of Triangular Lattice Spin Liquid Candidates," Chinese Phys. Lett. **35,** 117501 (2018).

[3] K. E. L. C. e. a. Bordelon, M.M., "Field-tunable quantum disordered ground state in the triangular-lattice antiferromagnet $NaYbO_2$," Nat. Phys. **15,** 1058–1064 (2019).

[4] M. M. B. et al., "Spin excitations in the frustrated triangular lattice antiferromagnet $NaYbO_2$," Physical Review B **101,** 224427 (2020).

[5] A. L. G. Joseph A M Paddison, J Ross Stewart, "SPINVERT: a program for refinement of paramagnetic diffuse scattering data," J. Phys.: Condens. Matter **25,** 454220 (2013).

[6] B. A. Frandsen, P. K. Hamilton, J. A. Christensen, E. Stubben, and S. J. L. Billinge, "*diffpy.mpdf*: open-source software for magnetic pair distribution function analysis," Journal of Applied Crystallography **55,** 1377–1382 (2022).