Final Technical Report

Non-Equilibrium Effects in Quantum Magnets

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Original PI: Kate A. Ross

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Abstract: While most often the state of a material will tend toward an equilibrium determined by its environment, there are many cases of scientific and technological interest where materials are manipulated to be or are found in non-equilibrium configurations. For example, data can be stored in hard drives by deliberately altering the magnetic orientation in a material to store information in non-equilibrium pattern. In this project, the main goals were studies of non-equilibrium properties of quantum magnets using neutron scattering as the primary experimental method. Neutron scattering allows characterization of magnetic correlation lengths sensitive to the presence of defects. It can also be used to distinguish equilibrium from non-equilibrium states via energy transfer rates. Typical bulk state magnetization relaxation times are too short to perform many neutron scattering measurements of interest. To enable the study of non-equilibrium conditions, materials with longer magnetic relaxation times were targeted. $CoNb_2O_6$ was used in two experiments related to non-equilibrium physics. In the first, evidence for defects created via the Kibble-Zurek mechanism (KZM) was sought by quenching across a magnetic field-dependent phase transition. Somewhat unexpectedly, clear evidence for KZM-induced defects was absent. Additional measurements of $CoNb_2O_6$ were made to better characterize its crystal field and other properties to provide a better theoretical understanding to enable more effective non-equilibrium physics measurements. In another project, $LiHo_{0.45}Y_{0.55}F_4$ was used to compare a quantum annealing protocol to a thermal annealing one since magnetic fields can be used to control the thermal fluctuations in $LiHo_{0.45}Y_{0.55}F_4$. In addition, a new pulsed magnet power supply and new techniques were developed suitable for neutron scattering experimental environments to enable faster magnetic field changes for producing non-equilibrium conditions. The power supply developed for this project has wider technological applications in addition to faster magnetic field ramps.

The general goal of the research in this project was to investigate the generation of non-equilibrium states in bulk quantum magnets and measure their properties. Quantum magnets are magnetic materials in which effects such as strong quantum fluctuations or entanglement are significant [1]. Pursuing this general goal required the development of new experimental techniques to allow for sufficiently rapid changes in magnetic fields that are applied during neutron scattering experiments plus the characterization of materials suitable for the planned studies.

There were three initial aims of the project:

- Aim 1: searching for and measuring the defects produced by a rapid magnetic field induced "quench" across a quantum critical point (QCP) [1] and comparing that to predictions derived from the Kibble-Zurek [2,3] mechanism
- Aim 2: investigating microscopic magnetic correlation changes in materials that display quantum annealing [4,5] such as $LiHo_xY_{1-x}F_4$
- Aim 3: search for many-body localization in a candidate material ($CoNb_2O_6$) after a magnetic field quench by examining the inelastic neutron scattering spectrum and comparing it to predictions based on thermal equilibrium to see if non-equilibrium states persist due to localization.

The first and third of these aims require comparatively fast ramps of substantial magnetic fields applied to a sample in a neutron scattering facility such as Oak Ridge National Laboratory (ORNL). This in turn necessitated the development of a pulsed power supply to produce the desired magnetic fields. The general findings associated with each of the three aims listed above will be described in this report along with the outlook for future related measurements and studies. In addition, the status of the development of the pulsed power supply will be presented as well.

Aim 1: Kibble-Zurek scaling in a magnetic quench through a quantum critical point

When there is a rapid change in temperature or a non-thermal parameter through a continuous phase transition, topological defects are predicted to be created through a mechanism proposed by Kibble [2] and later refined by Zurek [5]. This Kibble-Zurek mechanism (KZM) predicts that the density of these defects should have a power-law scaling with the quench rate that depends on very general parameters of the equilibrium properties of the system [3,6]. The KZM has been studied in systems such as superfluid helium [7], ultracold atomic gases [8,9], and defects in multiferroic materials [10,11]. As part of this project, the ac susceptibility of $CoNb_2O_6$ was measured to determine the feasibility of measuring and studying the KZM in a bulk material system (such as [12]) in an attempt to add to the range of systems in which the KZM can be studied.

$CoNb_2O_6$ is a material that has properties that map onto a quasi-1D Ising model [13-15]. This model is amenable to theoretical study and so measurements in $CoNb_2O_6$ have a potential to be particularly comparable to predictions. The goal of the proposed work was to use neutron scattering to measure defect densities in this material through measuring magnetic correlation lengths as a function of magnetic field quench rates. This was to be done through measuring the widths of magnetic Bragg peaks in different reciprocal directions since those widths are a function of the defect density and the widths could be measured as a function of quench rate.

While there have been extensive studies of $CoNb_2O_6$ over several decades including studies characterizing its magnetic phases [16-22], less attention has been paid to behavior in a transverse magnetic field [23] that leads to a correspondence with a transverse field Ising Model (TFIM) [13-15] in the system. An initial study was conducted prior to the neutron scattering experiments. AC susceptibility measurements were conducted at low temperatures. A transverse magnetic field quench into a zero-field freezing transition was used to study the relaxation response across a phase boundary. This relaxation response was studied as a function of the quench rate and while the data that were obtained were qualitatively in agreement with the predicted power-law scaling, several features such as a better fit to a logarithmic rather than power law decay and observed lack of dependence on whether the phase boundary was crossed or not indicated no experimental evidence for a KZM effect in this system. This result is somewhat surprising given the predicted universality of the KZM. It is also indicative of other physics to be explored and that care needs to be taken in analyzing $CoNb_2O_6$ defect-related responses. This work is reported in [24]. Additional analysis indicates that unexpectedly precise alignment with respect to the applied magnetic field is needed to fully realize the effective 1D Ising model in this context. Further measurements of $CoNb_2O_6$ are presented in the section discussing aim 3 below.

Searching for KZM effects in $CoNb_2O_6$ through neutron scattering experiments benefits from the ability to apply fast magnetic field quench rates because the ability to do so increases the range of parameters that can be studied and enhances the size of experimental signals. As part of this project, a power supply was developed that will enhance future KZM neutron scattering measurements in $CoNb_2O_6$ and other materials.

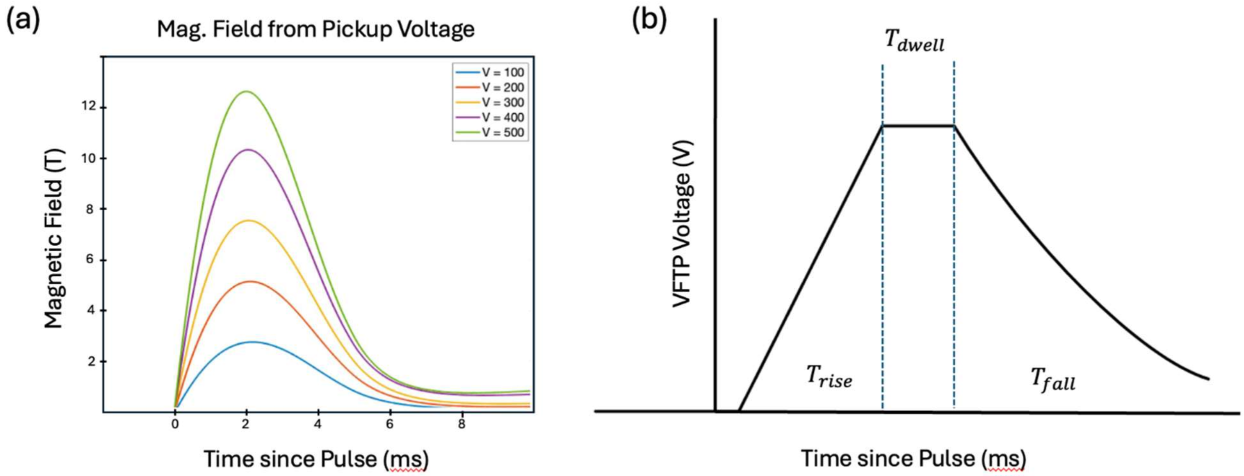

Fig. 1. (a) Maximum magnetic field strength as a function of time, on a coil fabricated manually. Displayed is an overall maximum field induction strength of approximately 12.6 T before the coil reasonably deteriorated due to high temperature from voltage and amperage loads. (b) Depiction of the VFTP main pulsing protocol, where the user determines how fast the field increases (T_{rise}), how long the sample is held at the maximum field (T_{dwell}), and how quickly the field discharges (T_{fall}).

This power supply was designed as part of this project because existing power supplies did not have the necessary specifications. A substantial magnetic field ramp rate whose rate can be adjusted over a wide range is necessary for the best realization of these experiments. The design called for a wide range of magnetic field ramp rates (0-10 T/s), as well as a large maximum field strength (approx. 12 T), to deal with the wide range of ramp rates needed to explore KZM as well as to explore the differing maximum field strengths needed to access the quantum critical point (QCP) in different TFIM materials. After work in partnership with ORNL [25], such a power supply that met these parameters was realized. See Figs. 1 and 2.

This device is called the Variable Fall Time Pulser (VFTP) for its ability to adjust magnetic field down-ramp rates across a wide range of such rates. It is not limited to powering magnets – its versatility extends to any pump-probe experiments in which variable ramp rates are needed. While its performance has been measured in test systems, it has not itself been tested in an experimental environment that fully corresponds to what is needed for use in a neutron scattering experiment. That work is continuing beyond the time period of this project.

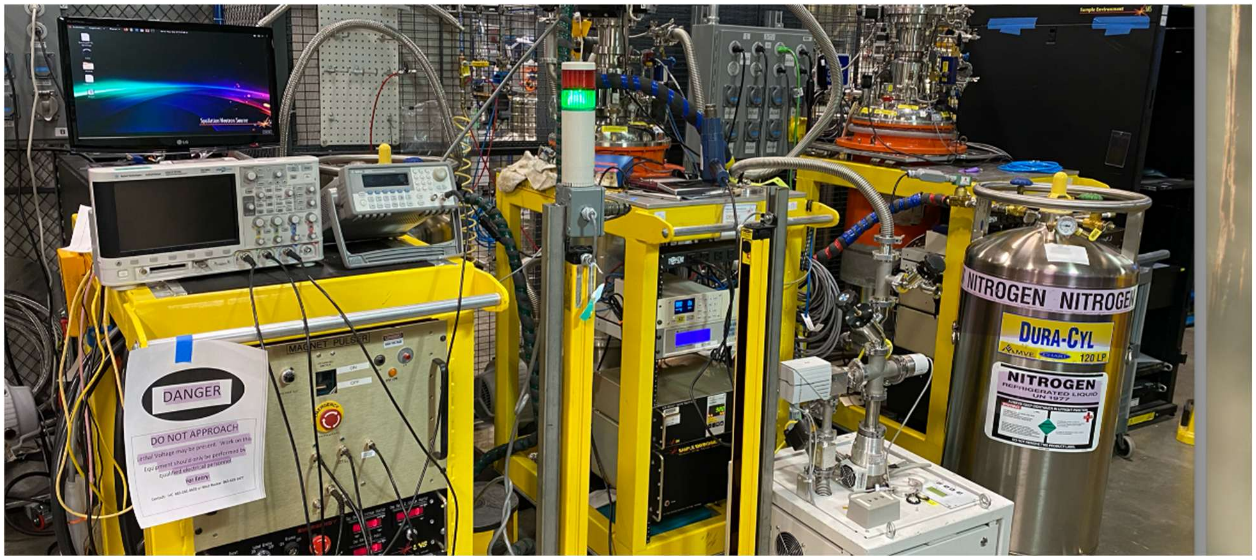

Fig. 2. Photograph of the VFTP in full non-experimental testing (including cryostat and magnet setup). Apparatus with "DANGER" indicator is the actual VFTP device.

Aim 2: Using Neutron Scattering to Study Quantum Annealing

Unlike aims 1 and 3 of this project, aim 2 does not require rapid magnetic-field quenches. Instead, techniques and materials like those used in aims 1 and 3 can be adapted to measuring quantum annealing [4,5]. For systems with a complicated energy dependence on system states, quantum tunneling can allow the system to minimize its free energy state more efficiently than through thermal annealing (heating and then cooling the material periodically to seek a minimum energy configuration to avoid cooling to a local rather than global minimum). Quantum annealing has applications in quantum computing [4] where it can be used to find the global minimum of a function in a time that can be much faster than classical methods.

Neutron scattering measurements were conducted in $LiHo_{0.45}Y_{0.55}F_4$, a material that can be described by a TFIM just as was the case for $CoNb_2O_6$ as described in aim 1. A transverse magnetic field was used to adjust the quantum fluctuations in this material to enable the comparison of a thermal annealing protocol with a quantum one that both started at the same initial and final temperatures and magnetic fields. By measuring the time evolution of spin correlations after the protocol there is evidence of greater change for the thermal annealing protocol, which would imply that the thermal annealing produced a state farther from equilibrium. However, measurements are also consistent with the presence of random magnetic fields produced by the applied transverse field, complicating the comparison of the two annealing methods. This work has been submitted for publication and is still under review as of the time of the submission of this report. The present data, analysis, and conclusions are available [26] and any further refinements will be reflected in the final version of the manuscript when published.

Aim 3: Many-Body Localization in Quantum Magnets

For materials with sufficient disorder, transport can be strongly inhibited. This leads to a Many-Body Localized (MBL) phase, which is a generalization of Anderson localization [27] physics to a system with interactions, after a quantum quench rather than an equilibrium phase. Given the link between $CoNb_2O_6$ and a TFIM description, rapid magnetic field quenches across phase boundaries can be used to search for MBL physics in this quantum magnet material [28] that has connections to theoretical predictions.

Partly in light of the observations described in aim 1, additional measurements of $CoNb_2O_6$ were performed to better understand deviations from the simplest Ising model description of the material when transverse magnetic fields were applied so that any measured results could be better interpreted. Crystal field levels of Co^{2+} were measured in pure $CoNb_2O_6$ directly using the SEQUOIA instrument at ORNL, which was reported in [29]. With the direct measurement of these states, we provided parameters for theorists to better investigate $CoNb_2O_6$ further as a candidate material for displaying MBL or MBL-like signatures.

Additionally, an effort was initiated to determine critical disorder levels in $CoNb_2O_6$ required to induce an MBL phase by synthesizing 7 variants of the system with different dilution strengths of Mg^{2+} in the compound, which serves to break up the Ising chains and vary the super exchange pathways and therefore better meet the requirements to display MBL. These samples were subjected to both neutron diffraction and magnetization measurements (on the POWGEN instrument at ORNL and MPMS3 *in situ* at Colorado State University), which so far seem to corroborate the idea that Mg^{2+} randomly replaces Co^{2+} in the Ising chains, but that is still a preliminary conclusion. Another preliminary finding is that small amounts of Mg^{2+} may stabilize the crystal structure. Data analysis was not completed prior to the conclusion of this project but is still ongoing with the intent of publishing these results in an upcoming student thesis [30]. Diffraction measurements allowed for a PDF (pair distribution function) analysis to determine crystal structure in order to make further crystal field calculations to confirm these preliminary conclusions.

Additional experiments conducted on both the CORELLI and GP-SANS instruments at ORNL where a magnetic field was ramped past a critical point and then back to zero to see if there were

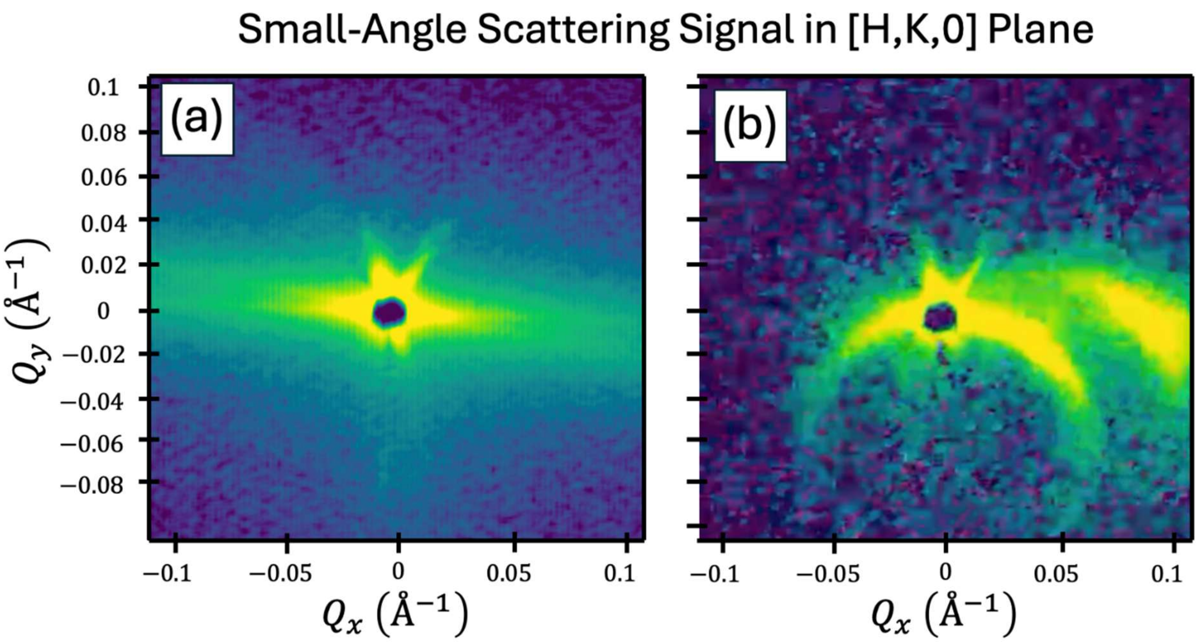

Fig. 3. Diffuse scattering in $CoNb_2O_6$. (a) Shows the scattering signal at zero field prior to ramping field upwards across the QCP. The crystal was likely misaligned in the ab-plane which accounts for the apparent second Bragg plane, but this detail is unimportant. (b) Scattering signal after the field crossed the QCP and relaxed to $B = 0T$. While the second signal to the right may indicate misalignment as stated previously, however, the unfamiliar circular diffuse structure is what is of interest in the current data analysis.

measurable impacts on the neutron scattering magnetic Bragg diffraction peaks. This was done to further explore the apparent glassy phase at low temperatures and magnetic fields reported in [24]. One observation from these measurements is that of an unusual diffuse scattering signal that indicates the presence of not-yet-understood physics for experimental signals and parameters similar to those that were to be used for KZ and MBL experiments. It is hoped that these unusual diffuse scattering results may lead to better theoretical understanding of the TFIM. Fig. 3 shows a neutron scattering signal that exhibits this diffuse scattering.

The fact that these experiments followed similar protocols as planned KZM and MBL experiments mean that there is a potential for any such better theoretical understanding to inform how best to observe these effects. An investigation of the relaxation dynamics and their origins in $CoNb_2O_6$ would likely be beneficial as well. Confirmation of any of these aims, such as the identification of an MBL phase and/or KZM defect production across a quantum phase transition, will undoubtedly require further exploration. Even so, it is likely that neutron scattering techniques will be an effective experimental approach and the ability to utilize a pulsed magnetic field with varying ramp rates such as the one developed in this project is advantageous for such efforts.

Summary

Neutron scattering measurements were conducted on materials that were amenable to a theoretical description based on a transverse field Ising model. Searches for both defects that were produced via the Kibble-Zurek Mechanism and Many-Body Localization signals were sought. Neither search produced unambiguous evidence of the effects of interest while revealing unanticipated and sometimes unexpected results. Additional studies provide information useful for better understanding the material used in these investigations, $CoNb_2O_6$, and indicating directions for future studies. In addition, an investigation into quantum vs. thermal annealing in $LiHo_{0.45}Y_{0.55}F_4$ was conducted where a transverse magnetic field was used to control the quantum fluctuations in the system, with neutron scattering again the primary experimental tool.

Authors of works associated with this project: E. S. Choi, A. I. Kolesnikov, G. Luke, H. S. Nair, T. R. Reeder, J. A. Ringler, S. Säubert, C. L. Sarkis, V. Williams, F. Ye, and K. A. Ross.

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