Machine Learning Assisted Insight to Spin Ice Dy$_2$Ti$_2$O$_7$

Anjana M Samarakoon* [1], Kipton Barros [2], Ying Wai Li [3], Markus Eisenbach [3,4], Qiang Zhang[1,6], Feng Ye [1], Z. L. Dun [5], Haidong Zhou [5], Santiago A. Grigera [7,8] , Cristian D. Batista [1,5], and D. Alan Tennant [4]

[1] Neutron Scattering Division, Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge TN 37831
[2] Theoretical Division and CNLS, Los Alamos National Laboratory, Los Alamos, NM 87545
[3] National Center for Computational Sciences, Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge TN 37831
[4] Materials Science and Technology Division, Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge TN 37831
[5] Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996
[6] Department of Physics and Astronomy, Louisiana State University, Baton Rouge, LA 70803
[7] Instituto de Física de Líquidos y Sistemas Biológicos, UNLP-CONICET, La Plata, Argentina
[8] School of Physics and Astronomy, University of St Andrews, United Kingdom.

*Corresponding author: samarakoonam@ornl.gov

(Dated: June, 2019)

Complex behavior poses challenges in extracting models from experiment. An example is spin liquid formation in frustrated magnets like Dy$_2$Ti$_2$O$_7$. Understanding has been hindered by issues including disorder, glass formation, and interpretation of scattering data. Here, we use a novel automated capability to extract model Hamiltonians from data, and to identify different magnetic regimes. This involves training an autoencoder to learn a compressed representation of three-dimensional diffuse scattering, over a wide range of spin Hamiltonians. The autoencoder finds optimal matches according to scattering and heat capacity data and provides confidence intervals. Validation tests indicate that our optimal Hamiltonian accurately predicts temperature and field dependence of both magnetic structure and magnetization, as well as glass formation and irreversibility in Dy$_2$Ti$_2$O$_7$. The autoencoder can also categorize different magnetic behaviors and eliminate background

---

Notice: This manuscript has been authored by UT-Battelle, LLC, under contract DE-AC05-00OR22725 with the US Department of Energy (DOE). The US government retains and the publisher, by accepting the article for publication, acknowledges that the US government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript, or allow others to do so, for US government purposes. DOE will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).
noise and artifacts in raw data. Our methodology is readily applicable to other materials and types of scattering problems.

Extracting the correct interactions from experimental data is essential for modelling. For magnetic insulators, the model is described by the spin Hamiltonian equation, dictated by symmetry, single-ion properties, and electron overlap between ions. The problem of extracting a spin Hamiltonian from neutron scattering data (inverse scattering problem) is often ill-posed and compounded by the need to use theory to interpret scattering data. Further, the available experimental data may not be enough to accurately determine the model parameters because of limited access to experimental data, a large noise magnitude at each scattering wavevector, or systematic errors associated with, e.g., background subtraction. Selecting the optimal Hamiltonian to model the experimental data is often a formidable task, especially when many parameters must be simultaneously determined. Tools for doing so are needed to uncover the new physics that is emerging from large classes of complex magnetic materials [1, 2]. Here we introduce a new autoencoder-based approach that can potentially address important modeling challenges, such as a proper background and noise subtraction, more reliable inference of model Hamiltonians, improved transferability to other physical systems, and efficiency.

Dy$_2$Ti$_2$O$_7$ is a highly frustrated magnet showing complex behavior including spin liquid formation [3, 4, 5, 6, 7, 8]. The magnetism originates from Dy$^{3+}$ ions which behave as classical Ising spins on a pyrochlore lattice of corner-linked tetrahedra, as in Fig. 1 (a). Figure 1 (b) shows the four essential magnetic interactions including a ferromagnetic coupling that results from the combination of exchange with a large dipolar interaction. This FM coupling makes Dy$_2$Ti$_2$O$_7$ a canonical spin-ice material, i.e., the spins on each tetrahedron obey the ice rules that only allow for two-in two-out configurations [9, 10]. This divergence free condition leads to a spin liquid with macroscopic degeneracy that features north and south charged magnetic monopoles interacting via a $1/r$ potential at elevated temperatures [3]. A full low-temperature characterization demands the study of a vast number of spins subject to short and long-range interactions. Spin dynamics occurs through millisecond quantum tunneling processes [11] and the measured characteristic equilibration time $\tau$ increases drastically leading to irreversible behavior below 600 mK [12, 13, 14, 15]. This slowdown has resulted in major difficulties [16, 17] in measuring and interpreting experiments such as heat capacity at low temperatures.
Here we use diffuse neutron scattering from time-of-flight techniques [see Methods: Experimental Details] on the CORELLI instrument at the Spallation Neutron Source, Oak Ridge National Laboratory to measure the magnetic state of Dy$_2$Ti$_2$O$_7$. Three dimensional (3D) volumes of diffuse scattering were measured in the 100mK to 960 mK temperature range. In view of the low temperature equilibration challenge, we undertake our analysis on data sets at 680 mK, which is low enough for correlations to be well developed but sufficiently high to reach equilibrium over a short time scale. Figure 2 (a) shows the background-subtracted data at 680 mK. This is proportional to the modulus squared of the spin components in the wavevector space. However, an additional aspect of neutron scattering is that it samples only the spin components perpendicular to the wavevector transfer $\mathbf{Q}$.

We employ a dipolar spin-ice Hamiltonian that includes exchange terms up to third-nearest neighbors:

$$
H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle \langle ij \rangle \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle \langle \langle ij \rangle \rangle \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_3' \sum_{\langle \langle \langle \langle ij \rangle \rangle \rangle \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D r_{ij}^3 \sum_{\langle ij \rangle} \left[ \frac{\mathbf{S}_i \cdot \mathbf{S}_j}{|r_{ij}|^3} - \frac{3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{|r_{ij}|^5} \right],
$$

where $\mathbf{S}_i$ can be viewed as an Ising spin of the $i^{th}$ ion, Fig. 1 (b). The model includes first, second, and two different third nearest neighbor interaction strengths, $J_1, J_2, J_3,$ and $J_3'$ respectively. There is also a dipolar interaction with strength $D$, which couples the $i^{th}$ and the $j^{th}$ spins, according to their displacement vector $\mathbf{r}_{ij}$. Prior work has determined $D = 1.3224$ K and $J_1 = 1.705$ K to high accuracy [17, 18, 19, 20, 21]. In the present modeling effort, we seek to determine the three unknown parameters $J_2, J_3, \text{ and } J_3'$ without any use of prior knowledge. Given a model Hamiltonian $H$, we use Metropolis Monte Carlo to generate a simulated structure factor, $S^{\text{sim}}(\mathbf{Q})$, to be compared with the experimental data $S^{\text{exp}}(\mathbf{Q})$ [Methods: Simulations].

In a direct approach, one might try to minimize the squared distance,

$$
\chi^2_S(\mathbf{Q}) = \frac{1}{N} \sum_{\mathbf{Q}} m(\mathbf{Q}) \left( S^{\text{exp}}(\mathbf{Q}) - S^{\text{sim}}(\mathbf{Q}) \right)^2,
$$

between the raw experiment and simulation data. We introduce a factor $m(\mathbf{Q}) \in \{0,1\}$ masking selected $\mathbf{Q}$-points where experimental artifacts can be identified [see Fig. S4]. The number of non-
masked $Q$-points is $N = \sum Q m(Q) \approx 1.2 \times 10^5$. We initially investigated optimization methods, such as the particle swarm method [22], to overcome local barriers and find the global minimum of $\chi^2_{S(Q)}$. However, despite nominal success in optimization, we quickly ran into reliability issues stemming from errors in the experimental and simulation data. As we will discuss below, $\chi^2_{S(Q)}$ is both noisy and effectively flat around its minimum, such that many distinct model Hamiltonians could achieve similarly small values of the $\chi^2_{S(Q)}$ error measure. Thus, even if we could find the global minimum of $\chi^2_{S(Q)}$, it might still be far from the physically correct model for Dy$_2$Ti$_2$O$_7$.

To address the ill-posed nature of this inverse scattering problem, we present two strategies: (1) We employ machine learning techniques to replace $\chi^2_{S(L)}$ with a new error measure $\chi^2_{S(L)}$ that is more robust to errors in the experimental and simulation data, and puts more weight on “characteristic features” of the structure factor. (2) Rather than reporting just the single “best” model, we sample from the entire set of Hamiltonian models for which the error measure is below some tolerance threshold. In this way, our method will report not just a model, but also a model uncertainty.

We use an autoencoder [23] to formulate $\chi^2_{S(L)}$, our new error measure. Autoencoders were originally developed in the context of computer vision, where they are known to be effective at image compression and denoising tasks. Here we apply them to interpret structure factor data, and to disambiguate among many possible solutions of the inverse scattering problem. Our autoencoder is a neural network that takes an $S(Q)$ as input (either simulated or experimental), encodes it into a compressed latent space representation $S_L$, and then decodes to an output $S_{AE}(Q)$ that captures the essence of the input $S(Q)$, while removing irrelevant noise and artifacts.

The autoencoder’s latent space $S_L = (S_1, S_2, \ldots, S_{30})$ provides a low-dimensional characterization of the $S(Q)$ data. Note that the physical $S(Q)$ data will contain many more scalar components than the 30 available in the latent space. Thus, by design, the autoencoder’s output $S_{AE}(Q)$ can only be an approximation to its input $S(Q)$. After proper training, one hopes that the autoencoder will be able to extract the relevant characteristics of a given $S(Q)$, while discarding irrelevant information.
such as noise and experimental artifacts. The autoencoder determines what information is relevant according to its ability to encode and faithfully decode the training data [Methods: Training].

We employ the simplest possible autoencoder architecture: a fully connected neural network with a single hidden layer. The hidden space activations (i.e., the latent space representation) are defined as

\[ S_L = f(\sum_Q W_{L,Q} m(Q) S(Q) + b_L), \]

where \( S(Q) \) is the input to the autoencoder, and the matrix \( W_{L,Q} \) and vector \( b_L \) are to be determined from the machine learning training process. Given simulated structure factor data as input, we interpolate to the experimental \( Q \)-points as necessary. The output of the autoencoder is defined as

\[ S_{AE}(Q) = f(\sum_{L=1}^{30} W'_{Q,L} S_L + b'_Q), \]

where the new matrix \( W'_{Q,L} \) and vector \( b'_Q \) are also trainable. We employ the logistic activation function \( f(x) = 1/(1 + e^{-x}) \) at both layers. This choice guarantees that the output \( S_{AE}(Q) \) is non-negative.

Figure 2 illustrates how the trained autoencoder processes the Dy\(_2\)Ti\(_2\)O\(_7\) scattering data. Figure 2 (a) shows the raw experimental data \( S^{\text{exp}}(Q) \) while Fig. 2 (b) shows how the autoencoder filters the experimental data to produce \( S^{\text{exp}}_{AE}(Q) \). The autoencoder preserves important qualitative features of the data, while being very effective at removing experimental artifacts. Figure 2 (c) shows the simulated data \( S^{\text{sim}}_{\text{opt}}(Q) \) for the optimal Hamiltonian model \( H_{\text{opt}} \), without any autoencoder filtering. We will describe later our procedure to determine \( H_{\text{opt}} \). Note that the best model, \( S^{\text{sim}}_{\text{opt}}(Q) \), is in remarkably good agreement with \( S^{\text{exp}}_{AE}(Q) \). This agreement is consistent with the fact that the autoencoder was trained specifically to reproduce simulated data.

Figure 3 provides another way to understand how the autoencoder is processing the \( S(Q) \) data. In Fig. 3(a) we show a cross section of \( S^{\text{exp}}(Q) \) in the high symmetry plane (\([h, -h, 0] - [k, k, -2k]\)). Figure 3(b) shows the corresponding simulated data \( S^{\text{sim}}_{\text{opt}}(Q) \) for the optimal model Hamiltonian. Now we perturb \( H_{\text{opt}} \) to a new model \( H_{\text{perturb}} \), which keeps all parameters from \( H_{\text{opt}} \) except modifies \( J_2 \) from 0.08 K to -0.09 K. Despite the relatively significant change to \( J_2 \), there is very little change to the structure factor. Indeed, Fig. 3 (c) shows that \( \Delta S_{\text{sim}} = S^{\text{sim}}_{\text{opt}}(Q) - S^{\text{sim}}_{\text{perturb}}(Q) \) is an order of magnitude smaller than the peaks in \( S^{\text{sim}}_{\text{opt}}(Q) \), and relatively noisy. This illustrates the inherent difficulty of our inverse scattering problem: many \( J_2 \) values seem to produce similarly good Hamiltonians.
The autoencoder latent space can be effective in extracting and amplifying important $S(Q)$ features that might otherwise be hidden. To show this, we consider the latent space representations $S_L$ and $S'_L$ for $S^\text{sim}_{\text{opt}}(Q)$ and $S^\text{sim}_{\text{perturb}}(Q)$, respectively. We ask: How much does $S_L$ need to be modified toward $S'_L$ in order to capture the important characteristics of $S^\text{sim}_{\text{perturb}}(Q)$, i.e., the perturbations to the structure factor? To answer this question, we replace 1, 6, and 12 latent space components of $S_L$ with the corresponding ones in $S'_L$. The components selected are those with the largest deviations, $|S_L - S'_L|$. Figures 3 (d), (e), and (f) show the change in autoencoder output, after substitution of the latent space components. Panel (f) reasonably captures the physically important characteristics of $\Delta S^\text{sim}$ while discarding irrelevant noise. Latent space components beyond 12 carry little information about $\Delta S^\text{sim}$.

Now we show how the autoencoder can assist in solving the inverse scattering problem, i.e., in finding the optimal model Hamiltonian $H_{\text{opt}}$ given the experimental data $S^\text{exp}(Q)$. To illustrate the important ideas, we first focus on determining $J_2$, assuming the other parameters of $H_{\text{opt}}$ are already known.

Figure 4 (a) shows $\chi^2_S(Q)$, as a function of $J_2$, illustrating the difficulty in making direct comparisons between experimental and simulated scattering data, Eq. (2). In principle the minimum of $\chi^2_S(Q)$ would give $J_2$, but in practice one must contend with relatively large uncertainties in the data. The visible scatter in Fig. 4 (a) is mostly a consequence of limited statistics of the simulated data. Other sources of error, such as systematic experimental error, will also exist and are more difficult to quantify.

A natural modification is to replace $\chi^2_S(Q)$ with the squared distance of autoencoder-filtered structure factors,

$$\chi^2_{S_{AE}}(Q) = \frac{1}{N} \sum_Q m(Q) \left( S^\text{exp}_{AE}(Q) - S^\text{sim}_{AE}(Q) \right)^2. \quad (3)$$

This new measure should be more robust to artifacts in both the experimental and simulation data. Indeed, as shown in Fig. 4 (b), it does slightly better in identifying an optimal $J_2$. 
Here we propose an alternative, and perhaps less obvious, error measure. The 30-dimensional latent space representation $S_L$ should, in some sense, capture the most relevant information in $S(Q)$. This suggests that to compare $S^{\text{exp}}(Q)$ to $S^{\text{sim}}(Q)$ we should actually look at the squared distance of their latent space vectors

$$\chi^2_{S_L} = \frac{1}{N_L} \sum_{L=1}^{30} \left( S^{\text{exp}}_L - S^{\text{sim}}_L \right)^2 . \quad (4)$$

Figure 4 (c) shows that this new error measure produces the clearest minimum, and thus the most precise identification of $J_2$. We will use $\chi^2_{S_L}$ as our optimization cost function in what follows.

The inverse scattering problem for Dy$_2$Ti$_2$O$_7$ requires finding not just one, but three unknown Hamiltonian parameters: $J_2$, $J_3$, and $J'_3$. We employ a variant of the Efficient Global Optimization algorithm to find the Hamiltonian $H_{\text{opt}}$ that minimizes $\chi^2_{S_L}$ [24]. In this approach, one iteratively constructs a dataset of carefully sampled Hamiltonians $H$. For each Hamiltonian $H$, we calculate a simulated structure factor $S^{\text{sim}}(Q)$ and corresponding deviation $\chi^2_{S_L}$ from the experimental data. With all such data, one builds a Gaussian process regression model $\hat{\chi}^2_{S_L}(H)$ that predicts $\chi^2_{S_L}$ for Hamiltonians $H$ not yet sampled. The low-cost model $\hat{\chi}^2_{S_L}(H)$ can be rapidly scanned over the space of Hamiltonians. Also, $\hat{\chi}^2_{S_L}(H)$ acts as a denoiser, effectively “averaging out” uncorrelated stochastic errors in the $\chi^2_{S_L}$ data. As more data is collected, the improved models $\hat{\chi}^2_{S_L}(H)$ will progressively become more faithful to $\chi^2_{S_L}$. Optimization, as described in methods [Methods: Optimization] gives the optimal parameters as $J_2 = 0.09(10)$ K, $J_3 = -0.002(25)$ K and $J'_3 = 0.051(9)$ K. The red curve of Fig. 4 (c) shows a cross section of the final $\hat{\chi}^2_{S_L}(H)$ model. The minimum at $J_2 = 0.08$ is readily apparent. The dashed line in Fig. 4 (a) indicates our empirically selected error tolerance threshold $C^2_{S(Q)}$. The dashed line in Fig. 4 (c) shows $C^2_{S_L}$, the corresponding tolerance threshold for the latent space error. We calculated $C^2_{S_L}$ from $C^2_{S(Q)}$ under the assumption of a fixed amount of uncertainty in the scattering data [Methods: Uncertainty]. Figure 4 (d) shows the three-dimensional regions of uncertainty corresponding to $\hat{\chi}^2_{S(Q)} < C^2_{S(Q)}$ (cyan) and $\chi^2_{S_L} < C^2_{S_L}$ (blue).
With more experimental constraints, we can further reduce uncertainty in $H_{\text{opt}}$. For this purpose, we define a new error measure $\chi^2_{\text{multi}} = \chi^2_{\hat{S}_L} \times \chi^2_{c_v}$, where $\chi^2_{c_v} = (c_v^{\text{exp}} - c_v^{\text{sim}})^2$ denotes the squared error between experimental [4] and simulated heat capacities, $c_v = \frac{1}{T^2} \langle U^2 - \langle U \rangle^2 \rangle$. Minimizing this multi-objective error function slightly modifies the model parameters: $J_2 = 0.00(3)$ K, $J_3 = -0.007(8)$ K and $J'_3 = 0.051(9)$ K, pictured as a green cross in Fig. 4 (d). But perhaps more importantly, the uncertainties in these parameters have decreased significantly. This is illustrated by the very compact dark-blue region in Fig. 4 (d), for which $\chi^2_{\text{multi}} < C^2_{\text{multi}}$, where $C^2_{\text{multi}}$ is again calculated as a function of $C^2_{\hat{S}_L}(Q)$.

The agreement between $S^{\text{exp}}(Q)$ and $S^{\text{opt}}_{\text{opt}}(Q)$ is quite good, as previously observed in Figs. 2 and 3. Further comparisons are shown in Fig. S2. To truly validate the model, however, we should compare to experimental data that has not been used during the model optimization process. For this purpose, we use the magnetic field dependence of different physical properties shown in Fig. 5. The optimal spin model reproduces the measured field dependence of the magnetization [25], the zero-field cooled (ZFC) and field cooled (FC) magnetic susceptibility [12], and the diffuse scattering at multiple temperature and applied field conditions, confirming that we have indeed found a model Hamiltonian adequate to describe the magnetic properties of Dy$_2$Ti$_2$O$_7$ including the onset of irreversibility and glassiness.

Our present study has primarily focused on robust inference of the optimal model Hamiltonian. There are two important aspects of our methodology that we wish to emphasize. First, our use of an autoencoder, trained on large quantities of simulation data, provides a distance measure $\chi^2_{\hat{S}_L}$ that allows robust comparisons to experimental scattering data. Second, our use of Gaussian process regression models $\hat{\chi}^2_{\hat{S}_L}$ as a low-cost predictor for $\chi^2_{\hat{S}_L}$ improves the quality of our optimized Hamiltonians. Gaussian process regression averages out uncorrelated stochastic error in $\chi^2_{\hat{S}_L}$, and helps in making uncertainty estimates. The latter is crucial for guiding the design of future experiments. Whereas traditional analysis of diffraction and inelastic neutron scattering is time consuming and error prone, our methodology is fully automated, and helps overcome difficulties of visualizing 3D or 4D data.
Finally, we remark that the autoencoder latent space provides an interesting characterization of structure factor data in its own right. Future studies might explore more direct application of autoencoders to the problems of background subtraction and of denoising experimental data. Here, we investigate another interesting application of the autoencoder: It can delineate different magnetic regimes. To demonstrate this, we will explore the space of $J_3$ and $J'_3$ parameters, while keeping $J_2 = 0$ K fixed. Our goal is to build a map of regimes with different dominant spatial magnetic correlations within this two-dimensional Hamiltonian space. We caution that the transitions between regimes will typically not be sharp phase transitions, so our modeling will not produce a phase diagram in the strict sense.

Figure 6 (a) shows the result of our clustering analysis on the simulated data [Methods: Clustering]. The optimal spin Hamiltonian for Dy$_2$Ti$_2$O$_7$ is marked as $H_{\text{opt}}$ near the center of this map. The corresponding $S_{\text{sim}}^r(Q)$ data, sliced in the high symmetry plane, had previously been shown in Fig. 3 (b). Figures 6 (b)-(i) show the $S_{\text{sim}}^r(Q)$ data for alternative Hamiltonians, as marked on the map. It is clear from these results that the spin Hamiltonian of Dy$_2$Ti$_2$O$_7$ is close to the confluence of multiple regimes. This fact reveals an additional source of complexity that explains the difficulties that were encountered in previous characterizations of this material. This analysis suggests a roadmap for further experimental studies. For example, the application of relatively small external fields and pressures or dopings should be enough to push Dy$_2$Ti$_2$O$_7$ into new magnetic regimes. For instance, the proximity to the ferromagnetic phase [blue regime in Fig. 6 (a)] indicates that the saturation field is small, as confirmed by magnetization, Fig. 5 (a).

In summary, a fundamental bottleneck in experimental condensed matter physics is model optimization and assessment of confidence levels. We have developed a machine learning-based approach that addresses both challenges in an automated way. Applied to Dy$_2$Ti$_2$O$_7$ our method produces a model that accounts for the diffuse scattering data as well as the lack of magnetic ordering at low temperature. Our approach readily extends to the analysis of dynamical correlations and other scattering data.
Methods

Experimental details

To measure the diffuse scattering of Dy₂Ti₂O₇ an isotopically enriched single crystal sample of Dy₂Ti₂O₇ was grown using an optical floating-zone method in a 5 atm oxygen atmosphere. Starting material Dy₂O₃ (94.4% Dy-162) and TiO₂ powder were first mixed in proper ratios and then annealed in air at 1400°C for 40 hours before growth in the image furnace as previously described [26]. Then the sample was further annealed in oxygen at 1400 °C for 20 hours after the floating zone growth. The lack of a nuclear spin moment in Dy-162 means that nuclear spin relaxation channels for the spins are cut off which is important in order to study the quench behavior in the material. In addition, the incoherent scattering from natural dysprosium is high (54.4 barns) whereas for Dy-162 it is zero and the absorption cross section is decreased from 994 barns (2200 m/s neutrons) for natural dysprosium to 194 barns for isotope 162. A best growth was achieved with a pulling speed of 3 mm/hour. One piece of crystal with the mass ≈ 200 mg was aligned in the (111) plane for the neutron investigation at the single crystal diffuse scattering spectrometer CORELLI at the Spallation Neutron Source, Oak Ridge National Laboratory. The crystal was prepared as a sphere to minimize absorption corrections and demagnetization corrections.

CORELLI is a time-of-flight instrument where the elastic contribution is separated by a pseudo-statistical chopper [27]. The crystal was rotated through 180 degrees with the step of 5 degree horizontally with the vertical angular coverage of ±8 degree (limited by the magnet vertical opening) for survey on the elastic and diffuse peaks in reciprocal space. The dilution refrigerator insert and cryomagnet were used to enable the measurements down 100 mK and fields up to 1.4 T. The data was reduced using Mantid [28] and Python script available at Corelli. Background runs at 1.4 Tesla were made to remove all diffuse signal and the extra scattering at Bragg peak positions due to the polarized spin contribution was accounted for by using the zero field intensities. [see Fig. S01] Fig. 2(a) and 3(a) shows a 3D plot and a slice of the high-symmetry plane of the background-subtracted diffuse scattering measurement at 680 mK and 0 T respectively.
Computational details

Simulations: Given a model Hamiltonian $H$, we use Metropolis Monte Carlo to generate a simulated structure factor, $S^{\text{sim}}(Q)$, to be compared with the experimental data $S^{\exp}(Q)$. We use simulated annealing to properly estimate $S^{\text{sim}}(Q)$ [29]. Beginning at an initial temperature of 50 K, we iterate through 11 exponentially spaced intermediate temperatures, until finally reaching the target temperature of 680 mK. At each intermediate temperature, $5 \times 10^6$ Monte-Carlo sweeps were performed. At every sweep, each spin is updated once on average, according to the Metropolis acceptance criterion [30]. We perform our simulations using $4 \times 4 \times 4$ cubic supercells, giving a total of 1024 spins in the pyrochlore lattice. The magnetic form factor of Dy$^{3+}$ and the neutron scattering polarization factor that enter in the calculation of the spin structure factor, $S(Q)$, are accounted before comparison to the background corrected experimental data. To correctly account for the long-range dipolar interactions, we used Ewald summation [31], implemented with the fast Fourier transform.

Training: To train the autoencoder, we require a dataset sufficiently broad to cover all potentially important characteristic features of the Dy$_2$Ti$_2$O$_7$ scattering data. For this purpose, we employ 1000 model Hamiltonians of the form Eq. (1). Each model has individually randomized coupling strengths $J_2, J_3, J_3'$, sampled uniformly from the range $-0.3$ K and $0.3$ K. For each model, we use simulated annealing to generate equilibrated three-dimensional $S^{\text{sim}}(Q)$ data at the target temperature of 680 mK. Our training data will thus consist of 1000 model Hamiltonians, each labeled by simulated data. The autoencoder tries to minimize the deviation between its input $S^{\text{sim}}(Q)$ and filtered output, summed over all random models in the dataset.

Training the autoencoder corresponds to determining the parameters (i.e., $W, b, W', \text{and } b'$) that minimize a loss function $\mathcal{L}$. Primarily, we are interested in minimizing the squared error between the simulated data and autoencoder-filtered output, summed over all models $H$ in the training dataset.
\[
\mathcal{L} = \sum_h \left[ \frac{1}{N} \sum Q \ m(Q) \left( S(Q) - S_{AE}(Q) \right)^2 + \mathcal{L}_R \right].
\]

The term \( \mathcal{L}_R \) is relatively weak, and includes two types of regularization: An \( L_2 \) regularization on the weight matrices \( W \) and \( W' \), and a sparsity regularization on the latent space activations \( S_L \) [32]. This regularization seems to improve the physical interpretability of the latent space representation. Despite having millions of trainable parameters in the neural network, the autoencoder does not seem prone to overfitting; the low-dimensionality of the latent space itself acts as a strong regularizer. To find the model parameters that minimize \( \mathcal{L} \), we use the scaled conjugate gradient descent algorithm [33], as it is implemented in Matlab. We also experimented with a Keras autoencoder implementation, and found that it made little qualitative difference in our final results.

**Optimization:** Optimization proceeds iteratively. We initially select 100 random Hamiltonians, where \( J_2, J_3, \) and \( J'_3 \) are each sampled uniformly from the range -0.3 K to 0.3 K. At each subsequent iteration, we use all available data to build \( \hat{\chi}^2_{S_L} \), the low-cost approximator to \( \chi^2_{S_L} \). Next, we randomly select 100 new Hamiltonians \( h \) for inclusion in the dataset, each being sampled uniformly, subject to the constraint \( \hat{\chi}^2(h) < c \). The cut-off parameter \( c \) decreases exponentially, rescaling by a factor 0.9 at each iteration. Consequently, later iterations in the optimization procedure are focused on regions where \( \chi^2_{S_L} \) is smallest. The optimization procedure terminates after about 40 iterations, at which point we take \( H_{opt} \) to be the minimizer of \( \hat{\chi}^2_{S_L}(H) \).

**Uncertainty:** How can we compare uncertainties of \( J_2 \), as estimated from \( \chi^2_{S}(Q) \) vs. \( \chi^2_{S_L} \)? From Fig. 4 (a) alone, one might estimate that \( J_2 \) could lie anywhere between \(-0.15 \) K and \( 0.25 \) K. This is the region for which \( \chi^2_{S}(Q) < C^2_{S}(Q) \), where \( C^2_{S}(Q) \) is an empirically selected tolerance denoted by the dashed horizontal line. Working backwards, we can then ask: How much noise in the simulated \( S^{\text{sim}}(Q) \) data would it take for \( C^2_{S}(Q) \) to be the actual stochastic uncertainty in \( \chi^2_{S}(Q) \)? Assuming that \( S^{\text{sim}}(Q) \) contains this level of noise magnitude, we can then measure the corresponding stochastic uncertainty \( C^2_{S_L} \) of \( \chi^2_{S_L} \), which we plot as the dashed line in Fig. 4 (c). Comparing with Fig. 4 (a), we conclude that the autoencoder-based error measure \( \chi^2_{S_L} \) is more robust to stochastic
noise, i.e., allows more precise estimation of $J_2$. Thus, we have selected $\chi^2_{S_L}$ as the best cost function for inferring the model Hamiltonian from the structure factor data.

Given experimental heat capacity data $c_p$, we introduced a multi-objective cost function $\chi^2_{\text{multi}}$. Repeating the same procedure as above, we can define the multi-objective tolerance threshold $C^2_{\text{multi}}$ in terms of the raw tolerance $C^2_S(Q)$.

**Clustering:** To determine magnetic regimes, we employ the agglomerative hierarchical clustering algorithm [34]. For this, we use the same dataset as was used to train the autoencoder, i.e., a random selection of 1000 model Hamiltonians, and their corresponding $S^{\text{sim}}(Q)$ data. The clustering algorithm requires as input the pairwise distances between all points in the dataset. We again employ the squared distance in the autoencoder latent space, i.e., as it appeared in $\chi^2_{S_L}$.

**Acknowledgements**

Z. L. D and H.D.Z thank the NSF for support with grant number DMR-1350002. A portion of this research used resources at Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. The research by D. A. T. was sponsored by the DOE Office of Science, Laboratory Directed Research and Development program (LDRD) of Oak Ridge National Laboratory, managed by UT-Battelle, LLC for the U.S. Department of Energy. (Project ID 9566). Support for Q. Z. was provided by US DOE under EPSCoR Grant No. DESC0012432 with additional support from the Louisiana board of regent. K. B. acknowledges support from the LDRD program at Los Alamos National Laboratory. Y.W.L., M.E., and resources for computer modeling are sponsored by the Oak Ridge Leadership Computing Facility, which is supported by the Office of Science of the U.S. Department of Energy under contract no. DE-AC05-00OR22725. D.A.T. and A.M.S. would like to thank Guannan Zhang for useful discussions.

**References**

[1] Gardner, J. S., Gingras, M. J., & Greedan, J. E., "Magnetic pyrochlore oxides.," *Reviews of Modern Physics*, vol. 82, no. 1, p. 53, 2010.

[2] D. Tennant, "Studies of Spinons, Majoranas, and Monopoles in Spin Liquid and Quantum Critical Magnets with Neutrons," *J. Phys. Soc. Jpn.*, vol. 88, p. 081009, 2019.

[3] Castelnovo, Claudio, Roderich Moessner, and Shivaji L. Sondhi., "Magnetic monopoles in spin ice," *Nature*, vol. 7174, no. 451, p. 42, 2008.
[4] Morris, D.J.P., Tennant, D.A., Grigera, S.A., Klemke, B., Castelnovo, C., Moessner, R., Czternasty, C., Meissner, M., Rule, K.C., Hoffmann, J.U. and Kiefer, K., "Dirac strings and magnetic monopoles in the spin ice Dy2Ti2O7.," *Science*, vol. 5951, no. 326, pp. 411-414, 2009.

[5] Fennell, Tom, P. P. Deen, A. R. Wildes, K. Schmalzl, D. Prabhakaran, A. T. Boothroyd, R. J. Aldus, D. F. McMorrow, and S. T. Bramwell, "Magnetic Coulomb phase in the spin ice Ho2Ti2O7.," *Science*, vol. 326, no. 5951, pp. 415-417, 2009.

[6] Henley, Christopher L, "The “Coulomb phase” in frustrated systems.," *Annu. Rev. Condens. Matter Phys.*, vol. 1, no. 1, pp. 179-210, 2010.

[7] Castelnovo, C., R. Moessner, and S. L. Sondhi, "Spin ice, fractionalization, and topological order.," *Annu. Rev. Condens. Matter Phys.*, vol. 3, no. 1, pp. 35-55, 2012.

[8] Rehn, J., and Roderich Moessner, "Maxwell electromagnetism as an emergent phenomenon in condensed matter.," *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*, vol. 374, no. 2075, p. 20160093, 2016.

[9] Harris, M. J., S. T. Bramwell, D. F. McMorrow, T. H. Zeiske, and K. W. Godfrey, "Geometrical frustration in the ferromagnetic pyrochlore Ho 2 Ti 2 O 7," *Physical Review Letters*, vol. 79, no. 13, p. 2554, 1997.

[10] Bramwell, Steven T., and Michel JP Gingras, "Spin ice state in frustrated magnetic pyrochlore materials.," *Science*, vol. 294, no. 5546, pp. 1495-1501, 2001.

[11] Castelnovo, C., R. Moessner, and S. L. Sondhi., "Thermal quenches in spin ice.," *Physical review letters*, vol. 104, no. 10, p. 107201, 2010.

[12] Snyder, J., B. G. Ueland, J. S. Slusky, H. Karunadasa, R. J. Cava, and P. Schiffer, "Low-temperature spin freezing in the Dy 2 Ti 2 O 7 spin ice," *Physical Review B*, vol. 69, no. 6, p. 064414, 2004.

[13] Snyder, J., J. S. Slusky, R. J. Cava, and P. Schiffer, "How ‘spin ice’freezes," *Nature*, vol. 413, no. 6851, p. 48, 2001.

[14] Jaubert, Ludovic DC, and Peter CW Holdsworth, "Magnetic monopole dynamics in spin ice," *Journal of Physics: Condensed Matter*, vol. 23, no. 16, p. 164222, 2011.

[15] Jaubert, Ludovic DC, and Peter CW Holdsworth., "Signature of magnetic monopole and Dirac string dynamics in spin ice.," *Nature Physics*, vol. 5, no. 4, p. 258, 2009.

[16] Melko, Roger G., Byron C. den Hertog, and Michel JP Gingras., "Long-range order at low temperatures in dipolar spin ice.," *Physical review letters*, vol. 87, no. 6, p. 067203, 2001.

[17] Borzi, Rodolfo Alberto, FA Gómez Albarracín, Héctor Diego Rosales, Gerardo Luis Rossini, Alexander Steppke, Dharmalingam Prabhakaran, A. P. Mackenzie, Daniel Carlos Cabra, and Santiago Andrés Grigera, "Intermediate magnetization state and competing orders in Dy 2 Ti 2 O 7 and Ho 2 Ti 2 O 7," *Nature communications*, vol. 7, p. 12592, 2016.

[18] Yavors’kii, T., Fennell, T., Gingras, M.J. and Bramwell, S.T., "Dy 2 Ti 2 O 7 spin ice: a test case for emergent clusters in a frustrated magnet," *Physical review letters*, vol. 101, no. 3, p. 037204, 2008.

[19] Henelius, P., Lin, T., Enjalran, M., Hao, Z., Rau, J.G., Altosaar, J., Flicker, F., Yavors'kii, T. and Gingras, M.J.P., "Refrustration and competing orders in the prototypical Dy 2 Ti 2 O 7 spin ice material.," *Physical Review B*, vol. 93, no. 2, p. 024402, 2016.
[20] Bovo, L., Twengström, M., Petrenko, O.A., Fennell, T., Gingras, M.J.P., Bramwell, S.T. and Henelius, P., "Special temperatures in frustrated ferromagnets.," Nature communications, vol. 9, no. 1, p. 1999, 2018.

[21] Giblin, S.R., Twengström, M., Bovo, L., Ruminy, M., Bartkowiak, M., Manuel, P., Andresen, J.C., Prabhakaran, D., Balakrishnan, G., Pomjakushina, E. and Paulsen, C., "Pauling Entropy, Metastability, and Equilibrium in Dy 2 Ti 2 O 7 Spin Ice.," Physical review letters, vol. 121, no. 6, p. 067202, 2018.

[22] Pérez, José Ignacio Hidalgo, Parallel Architectures and Bioinspired Algorithms, Heidelberg: Springer, 2012.

[23] Goodfellow, Ian, Yoshua Bengio, and Aaron Courville. , Deep learning, MIT press, 2016.

[24] Jones, Donald R., Matthias Schonlau, and William J. Welch, "Efficient global optimization of expensive black-box functions.," Journal of Global optimization, vol. 13, no. 4, pp. 455-492, 1998.

[25] Matsuhira, Kazuyuki, Zenji Hiroi, Takashi Tayama, Seishi Takagi, and Toshiro Sakakibara, "A new macroscopically degenerate ground state in the spin ice compound Dy2Ti2O7 under a magnetic field," Journal of Physics: Condensed Matter, vol. 14, no. 29, p. L559, 2002.

[26] Li, Q.J., Xu, L.M., Fan, C., Zhang, F.B., Lv, Y.Y., Ni, B., Zhao, Z.Y. and Sun, X.F, "Single crystal growth of the pyrochlores R2Ti2O7 (R= rare earth) by the optical floating-zone method.," Journal of Crystal Growth, vol. 377, pp. 96-100, 2013.

[27] Ye, Feng, Yaohua Liu, Ross Whitfield, Ray Osborn, and Stephan Rosenkranz, "Implementation of cross correlation for energy discrimination on the time-of-flight spectrometer CORELLI," Journal of applied crystallography, vol. 51, no. 2, pp. 315-322, 2018.

[28] Arnold, O., Bilheux, J.C., Borreguero, J.M., Buts, A., Campbell, S.I., Chapon, L., Doucet, M., Draper, N., Leal, R.F., Gigg, M.A. and Lynch, V.E, "Mantid—Data analysis and visualization package for neutron scattering and μ SR experiments," Nuclear Instruments and Methods in Physics Research, vol. 764, no. Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, pp. 156-166, 2014.

[29] Kirkpatrick, Scott, C. Daniel Gelatt, and Mario P. Vecchi., "Optimization by simulated annealing," science, vol. 220, no. 4598, pp. 671-680, 1983.

[30] Metropolis, Nicholas, Arianna W. Rosenbluth, Marshall N. Rosenbluth, Augusta H. Teller, and Edward Teller., "Equation of state calculations by fast computing machines.," The journal of chemical physics , vol. 21, no. 6, pp. 1087-1092, 1953.

[31] Rapaport, Dennis C., and Dennis C. Rapaport Rapaport. , The art of molecular dynamics simulation, Cambridge university press, 2004.

[32] Olshausen, Bruno A., David J. Field, "Sparse coding with an overcomplete basis set: A strategy employed by V1?,," Vision research, vol. 37, no. 23, pp. 3311-3325, 1997.

[33] M. F. Møller, "A scaled conjugate gradient algorithm for fast supervised learning.," Neural networks, vol. 6, no. 4, pp. 525-533, 1993.

[34] Zhang, Wei, Xiaogang Wang, Deli Zhao, and Xiaou Tang, "Graph degree linkage: Agglomerative clustering on a directed graph.," European Conference on Computer Vision, vol. Springer, no. Berlin, Heidelberg. pp. 428-441, 2012.
[35] Pomaranski, D., L. R. Yaraskavitch, S. Meng, K. A. Ross, H. M. L. Noad, H. A. Dabkowska, B. D. Gaulin, and J. B. Kycia. , "Absence of Pauling’s residual entropy in thermally equilibrated Dy 2 Ti 2 O 7.," Nature Physics, vol. 9, no. 6, p. 353, 2013.

[36] Moshtagh, Nima, "Minimum volume enclosing ellipsoid," Convex optimization, vol. 111, pp. 1-9, 2005.
Figure 1: (a) Atomic structure of Dy$_2$Ti$_2$O$_7$ comprised of tetrahedra of magnetic Dy ions (blue) and nonmagnetic octahedra of oxygen ions (red) surrounding Ti ions (cyan). (b) The magnetic moments located on Dy ions are constrained by crystal field interactions to point in or out of the tetrahedra. They form a corner sharing pyrochlore lattice. The pathways of nearest neighbor (1), next-nearest-neighbor (2) and two inequivalent next-next-nearest neighbor (3 and 3’) interactions are shown as thick colored lines.
Figure 2: Comparison of (a) the scattering function $S^{\text{exp}}(Q)$ for the Dy$_2$Ti$_2$O$_7$ crystal at 680mK, (b) the same experimental data after being filtered by the autoencoder, $S^{\text{exp}}_{\text{AE}}(Q)$ and (c), simulated scattering data from the optimal model spin Hamiltonian, $S^{\text{sim}}_{\text{opt}}(Q)$.

In going from (a) to (b), the autoencoder has filtered out experimental artifacts such as the red peaks, the missing data at the dark patches, etc. Using both scattering and heat capacity data, we determine the optimal spin Hamiltonian couplings to be $J_2 = 0.00(5)$ K, $J_3 = 0.005(10)$ K and $J_3' = 0.048(3)$ K with $J_1 = 1.705$ K and $D = 1.3224$ K having been fixed in prior work [17].
Figure 3: (a) Experimental scattering data $S^{\text{exp}}(Q)$ in the high symmetry plane. Dark patches indicate zero signal due to nuclear Bragg peaks. (b) The corresponding simulated data for the optimal model Hamiltonian. (c) The change in the simulated structure factor $\Delta S^{\text{sim}}$ when the $J_2$ coupling is perturbed from 0.08 K to -0.09 K. Note the change in color scale. The relatively weak response $\Delta S^{\text{sim}}$ to the large perturbation in $J_2$ illustrates the challenge in inferring the correct spin Hamiltonian. (d)-(f) With just 1, 6, and 12 latent space components, the autoencoder can reasonably capture important characteristics of $\Delta S^{\text{sim}}$. 
Figure 4: Inferring the spin Hamiltonian for Dy$_2$Ti$_2$O$_7$. (a) $\chi^2_{S(Q)}$ directly measures the distance between experimental and simulated $S(Q)$ data. It is relatively flat and noisy around its minimum, thus yielding a large uncertainty in the $J_2$ coupling (any value below the dashed line, $c^2_{S(Q)}$, is a reasonable candidate). (b) $\chi^2_{\text{AE}(Q)}$ measures the distance between $S(Q)$ data after being filtered through the autoencoder. (c) $\chi^2_{\text{SL}}$ measures the distance between the 30-dimensional latent space representations of the $S(Q)$ data. The Gaussian process model $\hat{\chi}^2_{\text{SL}}(H)$ (red curve) accurately approximates $\chi^2_{\text{SL}}$, even in the full space of $J_2$, $J_3$, and $J'_3$. (d) Once a good model $\hat{\chi}^2_{\text{SL}}(H)$ has been constructed, we can rapidly identify the optimal Hamiltonian model (magenta cross). The autoencoder-based error measure $\chi^2_{\text{SL}}$ yields much smaller model uncertainty (blue region) than the naïve one $\chi^2_{S(Q)}$ (cyan region). Model uncertainty is further reduced using a multi-objective error measure $\chi^2_{\text{multi}}$ that incorporates heat capacity data (dark blue region). Three most popular Hamiltonian sets currently used from ref. [17], ref. [18] and ref. [20] have also been marked as green, red and grey crosses respectively.
Figure 5: Validation of the optimal solution over multiple experiments: (a) Magnetization as a function of magnetic field and temperature, (b) zero-field cooled (ZFC) – field cooled (FC) susceptibility, magnetic diffuse scattering measured at 680 mK below (e) and at (f) the magnetization plateau. All the experiments and simulations shown here are done under magnetic field along the [1,1,1] direction. The magnetization and the ZFC-FC data are extracted from Refs. [26] and [5] respectively. All the experiments and simulations shown here are performed under magnetic field along [1,1,1] direction. The magnetization and ZFC-FC data are extracted from Refs. [26] and [5] respectively.
Figure 6: Map of regimes with different spatial magnetic correlations for varying $J_3$ and $J'_3$, with the remaining Hamiltonian parameters $J_1$, $J_2$ and $D$ being fixed to their optimal values. The colors in (a) indicate different groups of $S(Q)$, which correspond to regimes with different patterns of magnetic correlations. (b)-(i) Simulated $S(Q)$ data sliced in the high-symmetry plane at specific points indexed on panel (a). The large dark-blue regime (1) corresponds to long range ferromagnetic order, as indicated by the Bragg peaks in panel (b). (c)-(i) correspond to different patterns of short-range correlations arising from subsets of states that still obey ice rules. The pattern (g) falls within the same orange regime as $H_{opt}$, and qualitatively captures the Coulombic correlations expected for spin ice [cf. experimental scattering data in Fig. 3(a)].
Supplementary Information

Machine Learning assisted insight to the spin liquid material Dy$_2$Ti$_2$O$_7$

Anjana M Samarakoon, Kipton Barros, Ying Wai Li, Markus Eisenbach, Qiang Zhang, Feng Ye, Z. L. Dun, Haidong Zhou, Santiago A. Grigera, Cristian D. Batista, and D. Alan Tennant

This PDF file includes:

I. Additional information on experiment at CORELLI and data analysis
II. Heat Capacity data and simulation
III. Extra details on optimization and trained autoencoders
IV. Formulation of the optimal region

Figures S1-S6
I. Additional information on experiment at CORELLI and data analysis

The non-magnetic background for the scattering experiment at 680 mK was determined by replacing the Bragg peaks in raw data at 1.4 T with zero-magnetic field Bragg peaks. Fig. S1 (a) and (c) shows the raw data for 680 mK experiment at 0T and 1.4 T respectively.

Figure S1: Raw data from CORELLI experiment measured at 680 mK and magnetic field along [1,1,1] direction of 0 T (a) and 1.4 T (c) are shown here. The high-intensity rings are from the aluminum powder scattering from the sample holder and the instrument. The overall non-magnetic background (d) has been determined by replacing the Bragg peak intensity measured at 1.4 T by the zero-field values. The resulting background subtracted magnetic structure factor is shown in panel (b).
The slices $[k, k, -2k] - [l, l, l]$ and $[h, -h, 0] - [l, l, l]$ of the background subtracted data and best model simulations are shown in the Fig. S2.

Figure S2: Comparison between experimentally measured spin-spin correlations at 680mK (a)-(b) and modelling at the same temperature (c)-(d) along two perpendicular planes through $Q = 0$ in reciprocal space.
II. Heat capacity data and simulation

Figure S3: Specific Heat Capacity ($c_v$) data from ref. [4] and simulation at the optimized parameters given in the main text as a function of renormalized temperature. Below 600 mK (black dashed line), the temperature dependence of $c_v$ is controversial [35].
III. Extra details on optimization and trained autoencoders

Figure S4: Comparison between (a) raw, (b) background subtracted and (c) masked data at 680 mK. The instrumentation background was estimated as explained in Fig. S1. The mask was applied to background-subtracted data so as to exclude all the proposed measurement artifacts. The masked data was used to calculated $\chi^2_{S(I)}$ and $\chi^2_{AE}$ described in main text. (d), (e) and (f) are the autoencoder regularized structure factors, $S_{AE}(Q)$ for differently preprocessed input data (a), (b) and (c) respectively. Note that the axis limits of $S_{AE}(Q)$s are different from the original limits of $S^{exp}(Q)$ since the input datasets has been cropped to the same limits of $S^{sim}(Q)$ before regularizing through the trained autoencoder.
Figure S5: The extracted 30 features of $S(Q)$ along $[h, -h, 0] - [k, k, -2k]$ plane from the autoencoder trained with 1000 random samples in the three-dimensional parameter space.
Figure S6: The activation function along $J_3 - J_3'$ plane through $J_2 = 0$ for the 30 features shown on Fig. S5.
IV. Formulation of the optimal region

The three-dimensional optimal region for which $\chi^2_{\text{multi}} < c^2_{\text{multi}}$ as illustrated in Fig. 4(d) can be formulated by fitting the region to a minimum volume ellipsoid [36] as,

$$V \times [1.46 \quad -0.11 \quad 0.42] \times V^\dagger \leq 1; \quad V = [J_3 + 0.007 \quad J_3^' - 0.051 \quad J_2]$$

Note that, all the combinations of $J_2, J_3, J_3'$ which satisfies the above condition will reproduce both neutron structure factor and heat capacity behaviors.