Understanding temperature-dependent SU(3) spin dynamics in the $S = 1$ antiferromagnet $\text{Ba}_2\text{FeSi}_2\text{O}_7$

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Quantum magnets admit more than one classical limit and $N$-level systems with strong single-ion anisotropy are expected to be described by a classical approximation based on SU($N$) coherent states. Here we test this hypothesis by modeling finite temperature inelastic neutron scattering (INS) data of the effective spin-one antiferromagnet $\text{Ba}_2\text{FeSi}_2\text{O}_7$. The measured dynamic structure factor is calculated with a generalized Landau-Lifshitz dynamics for SU(3) spins. Unlike the traditional classical limit based on SU(2) coherent states, the results obtained with classical SU(3) spins are in good agreement with the measured temperature dependent spectrum. The SU(3) approach developed here provides a general framework to understand the broad class of materials comprising weakly coupled antiferromagnetic dimers, trimers, or tetramers, and magnets with strong single-ion anisotropy.

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INTRODUCTION

The computation of dynamical correlation functions at finite temperature is one of the important open problems of modern quantum many-body physics. These functions are not only crucial to test models against different spectroscopic techniques, but are also critical to the development of fast machine learning tools to accelerate and enhance understanding of problems at the forefront of condensed matter physics. For instance, the inelastic neutron scattering (INS) cross-section of quantum magnets is also critical to the development of fast machine learning tools to test models against different spectroscopic techniques, but are quantum many-body physics. These functions are not only crucial to the development of fast machine learning tools to accelerate and enhance understanding of problems at the forefront of condensed matter physics. For instance, the inelastic neutron scattering (INS) cross-section of quantum magnets is:

$$S(Q, E) = \sum_{\text{states}} \frac{\langle \psi_{\text{fin}} | \hat{S}_i^{(Q)} | \psi_{\text{init}} \rangle \langle \psi_{\text{init}} | \hat{S}_i^{(-Q)} | \psi_{\text{fin}} \rangle}{\Delta E}$$

Where $\hat{S}_i^{(Q)}$ is the dynamical spin structure factor, and $\psi_{\text{init}}$ and $\psi_{\text{fin}}$ are the initial and final states, respectively.

The main goal of this work is to test the aforementioned hypothesis by modeling the INS cross-section of the effective $S = 1$ quasi-2D easy-plane antiferromagnet (AFM), $\text{Ba}_2\text{FeSi}_2\text{O}_7$. In $\text{Ba}_2\text{FeSi}_2\text{O}_7$, a significant single-ion anisotropy ($D \sim 1.42$ meV) induced by the large tetragonal distortion of $\text{Fe}_2\text{O}_4$ tetrahedron in conjunction with spin-orbit coupling of $\text{Fe}^{2+}$ ($3d^5$) results in an effective low-energy three-level manifold generated by the spin states, $|S\rangle = |S = 0, \pm 1\rangle$. The competition with a relatively weak Heisenberg exchange interaction, places the ground state of this material ($a = J/D \sim 0.187$) near the quantum critical point at $c = 0.158$ that separates easy-planar AFM order from a quantum paramagnetic (QPM) phase (see Fig. 1). Since the AFM to QPM transition is driven by an enhancement of the local quadrupolar moment at the expense of the magnitude of the local dipolar moment, a proper classical description must allow for the coexistence of local dipolar and quadrupolar fluctuations, leading to transverse and longitudinal collective modes.$^{28}$ SU(3) coherent states fulfill this condition because an SU(3) spin has $8 = 3 + 5$
A quantum phase transition occurs above the sharp spin-waves disappear above $T_N$, the spectrum exhibits sharp spin-waves within the entire Brillouin zone (BZ) due to decay into a pair of transverse modes as described in ref. 28. While the sharp spin-waves disappear above $T_N$, a broad dispersion with a finite gap emerges at the magnetic zone center ($ZC$), $Q_{cl} = (1, 0, 0.5)$. With increasing temperature, the gap size increases and the bandwidth becomes narrower.

To understand the diffusive spectra above $T_N$, we performed a polarized neutron scattering experiment. Figure 2c shows the neutron spin polarization dependence of $I(Q,E)$ at $Q = (1, 0, 0)$. The spin-flip and non-spin-flip scattering cross-sections are coupled to the sample magnetization and the wave-vector, allowing us to extract the directional dependence of $S(Q,E)$. The neutron spin was polarized along $[0,1,0]$, which provides separate in-plane ($S^x = S^x(Q,E) + S^y(Q,E)$) and out-of-plane ($S^z = S^z(Q,E)$) components of $S(Q,E)$ for non-spin-flip and spin-flip channels, respectively. The non-spin-flip channel is for the most intense, indicating the diffusive spectra at 10 K mainly comes from the in-plane components of $S(Q,E)$.

**Calculation of generalized spin dynamics for SU(3) spin**

To account for the measured spectra at finite temperatures, we performed GSD calculations. The low-energy effective Hamiltonian for Ba$_2$FeSi$_2$O$_7$ is $\mathcal{H} = \sum_{r,s} \mathcal{O}^r_{\mu} \mathcal{O}^s_{\nu} + D \sum_r (\mathcal{O}^r_{\mu} + 2/3)^2$, with the convention of summation over repeated indices $\mu, \nu = x, y, z$ and $\mathcal{O}$ runs over the neighboring bonds with finite exchange interaction. This Hamiltonian can be recast in terms of SU(3) generators $\hat{O}^{\alpha}_{ij}$.

$$\mathcal{H} = \frac{1}{2} \sum_{r,s} \hat{O}^r_{\alpha} \hat{O}^s_{\alpha} + D \sum_r (\hat{O}^r_{\alpha} + 2/3)^2,$$

where the exchange tensor $\hat{J}^{\alpha}_{\beta} = \delta_{\alpha\beta}/J_0$, $\delta_{\alpha\beta}$, $\delta_{\alpha\beta}$ is the classical dipolar operators $S_i^{\alpha} S_j^{\beta}$. The neutron spin polarization dependence of $S(Q,E)$ at $Q = (1, 0, 0)$, the gap size decreases and the bandwidth becomes narrower. With increasing temperature, the gap size decreases and the bandwidth becomes narrower.
the generalized linear spin-wave calculation, the decay and renormalization of the $T_2$ and $L$-modes observed at 1.6 K (Fig. 2a) are not captured by this classical approximation. Capturing these feature requires the nonlinear approach described in ref. Above $T_N$, the GSD calculation reproduces the gapped nature of the spectrum representing a resonant excitation between $|S^z = 0\rangle$ and $|\pm 1\rangle$ states with a finite dispersion due to the exchange interaction. In the classical description, this diffusive mode originates from the combined effect of the "external SU(3) field" $D$ that induces a precession of each SU(3) moment with frequency $D/h$ (center of the peak) and the random molecular field due to the exchange interaction with the fluctuating neighboring moments that determines the width of the peak. When $J \ll T$, the spectrum thus becomes a dispersion-less broad peak centered around an energy $\Delta_{para} \approx D$. The computed spectra reproduce the main characteristics of the observed dispersions and bandwidth. Since the $|S^z = 0\rangle$ and $|\pm 1\rangle$ states are connected by the components that are transverse to the $z$-axis, $S^x = S^y \pm i S^z$, the corresponding intensity of $S(Q, E)$ should appear in the channel $S^x = S^y(Q, E) + S^z(Q, E)$ (Fig. 2d), which is qualitatively in good agreement with the polarization dependence of the measured $S(Q, E)$.

The detailed spectral change across $T_N$ is shown in Fig. 3 which compares the measured and calculated constant momentum scans at the ZC with varying temperature. For $T < T_N$, we consider a wave vector $Q = (1,0,0.2)$ that is close but not exactly equal to the ZC $Q = (1,0,0.5)$ in order to avoid the large tail of the magnetic Bragg reflection as well as the technical challenges associated with calculating the spectrum at the ZC. In this case, the $T_1$ (Goldstone) mode becomes visible because of its finite energy at $Q = (1,0,0.2)$ due to the non-zero $[0,0,L]$-dispersion produced by the small inter-layer coupling $\lambda_{inter}$. As a result, the three $T_1$, $T_2$, $L$ modes are observed in the spectrum (Fig. 3a). While the $T_1$ and $T_2$ transverse modes remain nearly unchanged with increasing temperature, the energy of the $L$-mode decreases and the mode becomes broader and indistinguishable from the quasielastic scattering near $T_N$. Above $T_N$, the quasielastic scattering continuously evolves into a broad peak centered at finite energy ($\Delta_{para}$), whose energy increases gradually with the temperature (see Fig. 3c). To extract the spectral weight of the resonant excitation above $T_N$ and the $L$-mode below $T_N$, the data were fitted with a double Lorentzian function associated with a damped harmonic-oscillator (DLDHO),

$$S(E) = A(n(E) + 1) \left[ \frac{\Gamma}{(E - \Delta)^2 + \Gamma^2} + \frac{\Gamma}{(E + \Delta)^2 + \Gamma^2} \right]$$

that provides a simplified description of the contribution of an over-damped mode. The $n(E) = 1$ is the Bose factor, and $A$, $\Delta$, and $\Gamma$ indicate the intensity, energy, and line-width of the peak, respectively. The extracted spectral weights and parameters of the $L$-mode and the resonant excitations are indicated by the shaded regions in Fig. 3a–c, and summarized in Fig. 3d, e, respectively.

Figure 3b, c show a comparison of the GSD calculation with the INS data across $T_N$. Remarkably, the spectral weight for the $L$-mode is enhanced and shifts to low-energy with increasing temperature, which is consistent with the data. Above $T_N$, the GSD calculation gives a diffusive resonant peak-shaped spectrum centered at the energy $\Delta_{para}$ that approaches $D$ for $T \gg T_N$. We note that the traditional LLD based on SU(2) coherent states cannot explain this gapped diffusive mode as well as the $L$-mode, leading to incorrect results in the high-temperature limit. However, we notice that the calculated spectrum underestimates the width of the mode at temperatures $T \approx T_N$. This discrepancy arises from an inadequate renormalization of the SU(3) spins at $T \approx T_N$, similar to the issue raised in traditional SU(2) LLD. This discrepancy in the line-width can be removed in the high-temperature limit by applying an adequate renormalization of the SU(3) spins $q_{苏} \rightarrow \kappa q_{苏}$, with $\kappa = 2$ in the high-temperature ($T \gg T_N$) limit, as described in ref. This renormalization guarantees that the SU(3) $S(Q, E)$ satisfies the exact sum-rule in the high-$T$ limit. Properly renormalizing the spin has the additional virtue of bringing the theoretical Néel temperature to $T_{N}^{\text{eff}} \approx 7.5 K$ ($\kappa = 2$) closer to the experimental value $T_{N} = 5.2 K$. Figure 4a shows the comparison between the new calculations including the renormalization factor and the data measured at the same temperature $T \approx T$ (experiment). This comparison reveals a better agreement in spectral shape at $T_N < T$ than those without renormalization.
A crucial advantage of the GSD demonstrated here is that the direct problem (inferring the INS cross-section of a given scattering problem) requires the development of fast solvers of extraction models from data, the solution of this important consequences for the characterization of quantum limiting cases $\kappa$ function.

The transverse and longitudinal modes are indicated with labels 'T$_N$', 'T$_Z$' and 'L'. The spectral weight for the L-mode was fitted with a DLDHO function, and the results are indicated by the orange (blue) shaded regions for experimental (calculated) spectra. The calculated intensities by the GSD are shown in b, c. Comparison of the measured and calculated momentum scans at the ZC above $T_N$. As in a, b, the spectral weights were determined by fitting with a DLDHO function and the results are indicated by the shaded regions. The extracted spectral shapes of the resonant modes are compared with the GSD calculations shown by the blue solid line. d, e Comparison of contour plots of the constant momentum scans between INS data and GSD calculation across $T_N$ and $T_N^2$. The position ($\Delta$) and line-width ($\Gamma$) of the L-mode and resonant excitation were quantified by the DLDHO function, and are exhibited as points with error-bars, respectively.

DISCUSSION
In summary, the GSD based on SU(3) spin provides a good approximation of the measured INS cross-section over a broad temperature range. The most quantitative deviations are observed at very low-temperatures $T \ll T_N$ and close to $T_N$. The former case is due to the requirement of a one-loop quantum correction to account for the decay of the L-mode, and the latter is due to the expected discrepancy between the experimental and re-scaled values of $T_N$ originated from the renormalization factor $\kappa = 2$ to the classical SU(3) spins. This renormalization factor arises from enforcing the sum-rule in the infinite T-limit. Similarly, the GSD of unrenormalized classical SU(3) ($\kappa = 1$) leads to the correct sum-rule in the zero temperature limit after quantizing the normal modes. Therefore, the correct scaling factor should be defined as a function $\kappa(T)$ that monotonically interpolates between the two limiting cases $\kappa(0) = 1$ and $\kappa(\infty) = 2$.

The verification of the main hypothesis of this work has very important consequences for the characterization of quantum magnets. For instance, while INS is an ideally suited technique for extracting models from data, the solution of this “inverse scattering problem” requires the development of fast solvers of the direct problem (inferring the INS cross-section of a given model). A crucial advantage of the GSD demonstrated here is that the cost of the simulations scales linearly in the system size, while the computation cost of exact dynamics grows exponentially, making it an ideal solver for attacking the inverse scattering problem with machine-learning-based approaches. Moreover, since the GSD can reproduce the INS data in the high-temperature regime, the method can still be applied to quantum magnets that exhibit long-range entanglement at low enough temperatures, but undergo a quantum to classical crossover above a certain temperature $T_{Qc}$. Finally, we note that the SU(N) approach described here is also relevant to the broad class of materials comprising weakly coupled antiferromagnetic magnets including dimers, trimers, or tetramers as well as magnets with strong single-ion anisotropy, where similar effects may be anticipated.

METHODS
Inelastic neutron scattering experiment
For the INS experiments, the same single crystal of Ba$_2$FeSi$_2$O$_7$ (mass: 2.13 g) as was used in ref. was aligned on an aluminum plate with an [H, 0, L] horizontal scattering plane. Unpolarized INS data were collected using the cold neutron triple-axis spectrometer (CTAX) at the High Flux Isotope Reactor (HFIR) at the Spallation Neutron Source (SNS) located at Oak Ridge National Laboratory. A liquid helium cryostat was used to control temperature for both experiments. At CTAX, the initial neutron energy was selected using a PG (002) monochromator, and the final neutron energy was fixed to $E_i = 3.0$ meV by a PG (002) analyzer. The horizontal collimation was guide - open - 40° - 120°, which provides an energy resolution with full width half maximum (FWHM) = 0.1 and 0.18 meV for $E = 0$ and 2.5 meV, respectively. For the HYSPEC experiment, $E_i = 9$ meV and a Fermi chopper frequency of 300 Hz were used, which gives FWHM = 0.28 meV and 0.19 meV of energy resolution at $E = 0$ and 2.5 meV, respectively. Measurements were performed by rotating the sample from −50° to 170° with 1° steps. Data were integrated over $K = [-0.16, 0.16]$ and $L = [L - 0.1, L + 0.1]$, and symmetrized over positive and negative $H$.
We also performed polarized neutron scattering measurement as part of the HYSPEC experiment using XYZ-polarization analysis which is the same configuration as the experiment in ref. 40. The X-axis is defined along \( Q = [1, 0, 0] \) for the scattering wave vector, the Z-axis is defined along \([0, 0, 1]\) perpendicular to the scattering plane, and the Y-axis is defined along the direction \([0, 0, 1]\) perpendicular to the X- and Z-axes. In the experiment, the neutron was polarized along the Z-direction, and the nonspin-flip and spin-flip scattering cross-sections provide \( I_{0}(E) + I_{mag}^{X}(E) \) and \( I_{mag}^{Y}(E) \), respectively. The \( I_{0}(E) \) indicates non-magnetic structure factor and \( I_{mag}^{X}(E) \), where \( \alpha \in \{X, Y, Z\} \), is the component dependent magnetic structure factor. The measured spin-flip and nonspin-flip cross-sections provide distinct \( I_{mag}^{X}(E) \) and \( I_{mag}^{Y}(E) \), which correspond to \( S^{|x|} + 2S^{|y|} \) and \( S^{|z|} \), respectively, on the crystal axis where \( x|a, y|b, \) and \( z|c \) in the tetragonal crystal structure. All of data sets were reduced and analyzed using the MANTID\(^{42}\) and DAVE\(^{43}\) software packages.

**DATA AVAILABILITY**

Data are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

S.H.D., H.Z., C.D.B., and A.D.C. conceived the project. T.H.J., S.W.C., and J-H.P. provided single crystals. S.H.D., T.J.W., T.H., V.O.G., and A.D.C. performed INS experiments. S.H.D., and A.D.C. analyzed the neutron data. H.Z., D.A.D, K.B., and C.D.B. constructed theoretical model and calculations. S.H.D., H.Z., C.D.B., and A.D.C. wrote the manuscript with input from all authors.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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