Dynamical Scaling as a Signature of Multiple Phase Competition in $Yb_2Ti_2O_7$

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$Yb_2Ti_2O_7$ is a celebrated example of a pyrochlore magnet with highly frustrated, anisotropic exchange interactions. To date, attention has largely focused on its unusual, static properties, many of which can be understood as coming from the competition between different types of magnetic order. Here we use inelastic neutron scattering with exceptionally high energy resolution to explore the dynamical properties of $Yb_2Ti_2O_7$. We find that spin correlations exhibit dynamical scaling, analogous to behavior found near to a quantum critical point. We show that the observed scaling collapse can be explained within a phenomenological theory of multiple-phase competition, and confirm that a scaling collapse is also seen in semiclassical simulations of a microscopic model of $Yb_2Ti_2O_7$. These results suggest that dynamical scaling may be general to systems with competing ground states.

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Frustration generates competition. When the interactions of a many-body system are frustrated, it is common to find many competing phases close in energy to the ground state [1–3]. Even though some particular order may emerge as the stable ground state at sufficiently low temperature, the proximity of the competing phases may still have a substantial influence on the system's properties [4–9]. In such a case, we must understand the system through the lens of multiple phase competition.

This multiple phase competition perspective has yielded especially helpful insight into rare-earth pyrochlore magnets [7], most prominently $Yb_2Ti_2O_7$ [7,10–33]. Composed of magnetic Yb^{3+} ions arranged in a lattice of corner-sharing tetrahedra, the system orders ferromagnetically at $T = 270$ mK [27,34,35]. Its magnetic Hamiltonian lies extremely close to the boundary between canted ferromagnetic (FM) order and Γ_5 antiferromagnetic (AFM) order [17,22,26,32]. And in the broader parameter space, this phase boundary terminates in a spin liquid where it meets a Palmer-Chalker AFM [7,36]. Various static properties of $Yb_2Ti_2O_7$, such as its low ordering temperature, the strong variation between samples and the equal-time spin correlations have been understood as arising from multiple phase competition [4,7,22,23,26,32].

However, not all behaviors of $Yb_2Ti_2O_7$ are well understood, particularly those relating to dynamics. Above the long range magnetic ordering transition $T = 270$ mK and up to $T \sim 2$ K, $Yb_2Ti_2O_7$ is in a short-range correlated magnetic phase [10]. In this temperature regime, diffuse rods of neutron scattering appear along the $\{111\}$ directions, which signal structured spin correlations [14–16,18,22]. The presence of these rods is a signature of the proximity of AFM order, and thus falls within the picture of multiple phase competition, but their energy dependence remains an open issue. Meanwhile, thermal conductivity [37,38] and thermal hall conductivity [39] reach anomalously large values in the $Yb_2Ti_2O_7$ short-range correlated phase, and terahertz spectroscopy appears to show the presence of massive magnetic quasiparticles [40]. Thus the $0.27\ \text{K} < T < 2$ K magnetic state hosts exotic but poorly understood dynamics. This raises the question, can the intermediate temperature dynamics of $Yb_2Ti_2O_7$ be understood via multiphase competition? What role, if any, does the nearby spin liquid play? And, more generally, does multiple-phase competition imply anything universal about the dynamics of the disordered phase, in analogy with quantum criticality?

In this study, we experimentally demonstrate a universal scaling relation for the energy dependence of the rodlike scattering in $Yb_2Ti_2O_7$ and connect it with the multiple phase competition paradigm. This is accomplished through low-energy neutron scattering measurements between 0.3 and 2 K, using ultrahigh resolution inelastic neutron

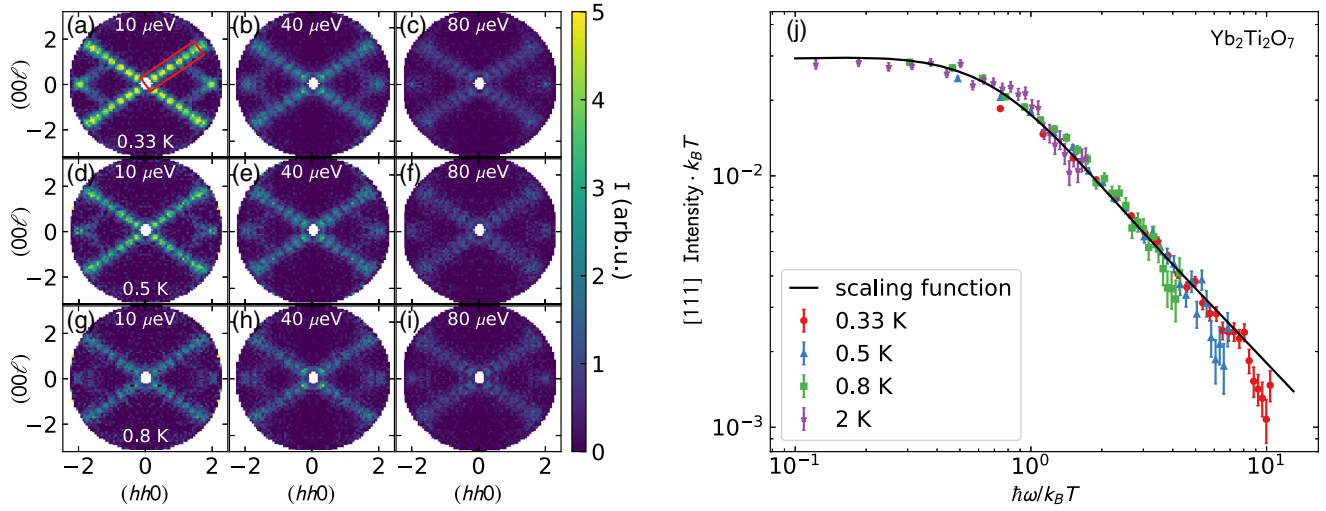

FIG. 1. Low-energy neutron scattering from the short-range correlated phase of $Yb_2Ti_2O_7$. Panels (a)–(i) show color plots of neutron scattered intensity, with the horizontal rows showing three different temperatures and the vertical columns show different constant energy slices in the $hh\ell$ scattering planes. All temperatures and energies show diffuse scattering rods along $\{111\}$ directions along with crosses of scattering centered at (220). Panel (j) shows the data integrated over the $\{111\}$ scattering rods [indicated by the red box in panel (a)] scaled by the temperature. Up to 2 K, the data collapse onto themselves and follow a scaling relation of type Eq. (1). The specific form of scaling predicted by our phenomenological theory of multiple-phase competition, Eq. (3), is shown with a solid line. Error bars indicate 1 standard deviation.

spectroscopy. The inelastic neutron scattering intensity along the $\{111\}$ directions of reciprocal space, $S_{\rm rod}(\omega)$, is described by a scaling relation (derived below) between temperature and energy, reminiscent of scaling laws observed in quantum critical systems [41–45]:

$$k_B T S_{\rm rod}(\omega) = f\left(\frac{\hbar\omega}{k_B T}\right). \tag{1}$$

We then show how this scaling relation can be understood within a phenomenological theory of multiple phase competition, combined with Langevin dynamics. This theory is corroborated using semiclassical molecular dynamics simulations of a microscopic model known to describe $Yb_2Ti_2O_7$, which confirm that the scaling behavior is associated with the region of parameter space where FM-AFM orders compete. We thus show that multiple phase competition has universal consequences—independent of the precise Hamiltonian—in finite-temperature dynamics.

We measured the low-energy inelastic neutron spectrum of $Yb_2Ti_2O_7$ between 0.3 and 2 K using the ultrahigh resolution BASIS backscattering spectrometer [46] at ORNL's SNS [47]. The sample was two stoichiometric single crystals grown with the traveling solvent floating zone method [34] (the same crystals as Ref. [32]) co-aligned in the $(hh\ell)$ scattering plane, and mounted in a dilution refrigerator. We rotated the sample over 180° about the vertical axis, measuring the scattering up to 300 μeV (the full bandwidth of this configuration) with 3 μeV full width at half maximum energy resolution—much higher resolution than previous measurements of these features. Constant-energy slices of the data are shown in Fig. 1. We measured the $Yb_2Ti_2O_7$ spectrum at temperatures 330, 500, and 800 mK with 12 K background in one experiment, and then 330 mK, 2 K, 3 K, with 12 K background in a second experiment with the same sample. (12 K is well into the paramagnetic phase where all spin correlations are lost, and thus makes an appropriate background for the inelastic data—see Supplemental Material for details [48].) Because of beam heating, the cryostat thermometer may differ from the actual sample temperature; accordingly, the temperature of the lowest temperature measurement (for which the cryostat thermometer temperature was reading below 100 mK) was derived from a fitted Boltzmann factor (detailed balance) for the positive and negative energy transfer scattering on the $\{111\}$ feature: $T = 0.33(4)$ K.

As is clear from Fig. 1, the inelastic scattering pattern in the short-range correlated phase has well-defined rods of scattering extending along the $\{111\}$ directions. As energy transfer ω increases, the scattering pattern grows weaker and broadens, but does not change its overall character. Intriguingly, the same effect is observed as temperature increases: the rod scattering pattern is preserved but grows weaker and broader. This raises the question of whether there is a scaling relation between temperature and energy.

To test this hypothesis, we integrated the $\{111\}$ rod scattering [shown by a red box in Fig. 1(a)] and plotted the intensity multiplied by temperature as a function of energy divided by temperature in Fig. 1(j). We find that the data collapse onto a universal curve, and above $\hbar\omega/k_B T \approx 1$ the data follow a $(\hbar\omega/k_B T)^{-n}$ power law behavior, with a fitted exponent $n = 1.03(3)$. (In the Supplemental Material,

we show this exponent to be robust against different Q integration regions [48].) This implies a scale invariance in the dynamics of the $Yb_2Ti_2O_7$ short-range correlated phase.

To understand this, we construct a phenomenological theory that takes into account the competition between FM-AFM phases. Writing a Ginzburg-Landau theory with dissipative dynamics [49] in terms of competing order parameters of ferromagnetic and antiferromagnetic phases, and assuming low-energy modes along $\{111\}$ that collapse to zero energy at some temperature $T_{\rm rod}$, we find an equation (derived in the Supplemental Material [48]) for the inelastic structure factor of a [111] rod $S_{\rm rod}$

$$S_{\rm rod}(\omega) = \int_{q_1}^{q_2} S(q_{111}, \omega) = 2(n_{BE}(\omega)+1)\frac{1}{k_B^2(T-T_{\rm rod})^2}\frac{B\hbar\omega}{R^2+\frac{\hbar^2\omega^2}{k_B^2(T-T_{\rm rod})^2}}. \quad (2)$$

Here B and R are nonuniversal dimensionless constants, and $n_{BE}(\omega)$ is the Bose-Einstein distribution.

Fitting Eq. (2) to the $Yb_2Ti_2O_7$ experimental data, we find good agreement with $T_{\rm rod} = -0.05(5)$. This is zero to within uncertainty. Setting $T_{\rm rod} = 0$ explicitly we obtain the scaling relation (1), with the scaling function

$$f(x) = 2\left(\frac{1}{\exp(x)-1}+1\right)\frac{Bx}{R^2+x^2}, \quad (3)$$

which depends only on the ratio $x = (\hbar\omega/k_BT)$. This form for $f(\hbar\omega/k_BT)$ beautifully matches the experimental data as shown in Fig. 1, with fitted constants $B = 0.0181(3)$ and $R = 0.80(3)$. (The high $\hbar\omega/k_BT$ data slightly deviates from the scaling law; in the Supplemental Material we discuss how this is likely a background subtraction artifact [48].)

The crucial ingredients in the phenomenological theory behind Eq. (3) are (i) dissipative dynamics; (ii) close competition between two phases, here ferromagnetic and antiferromagnetic; (iii) flat, low energy modes, along the $\{111\}$ directions; (iv) a collapse of these modes to zero energy at some temperature $T_{\rm rod}$; (v) $T_{\rm rod} \approx 0$.

Of these, (i) is a natural assumption for a paramagnetic phase in a strongly interacting system, (ii) has been inferred previously from the static behavior of $Yb_2Ti_2O_7$ [22,23,32] and (iii) is known to follow from (ii) [7] with $\{111\}$ being the direction of the low energy modes arising from the microscopic theory [48]. Explaining the data then requires one novel assumption [(iv)] and an empirical determination that $T_{\rm rod} \approx 0$ for $Yb_2Ti_2O_7$ [48].

To validate the idea of a temperature-dependent, collapsing, energy scale for the $\{111\}$ rods in a microscopic model appropriate to $Yb_2Ti_2O_7$, we turn to molecular dynamics simulations. We simulate a nearest-neighbor anisotropic exchange Hamiltonian

$$H_{\rm ex} = \sum_{\langle ij\rangle}\sum_{\alpha\beta} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta. \quad (4)$$

The form of the exchange matrices $J_{ij}^{\alpha\beta}$ is fixed by symmetry [7,17,50] and there are four independent parameters $\{J_k\} = \{J_1, J_2, J_3, J_4\}$. Several different estimates of these parameters are available for $Yb_2Ti_2O_7$ [17,22,26,32], generally placing $Yb_2Ti_2O_7$ close to a phase boundary between ferromagnetic and antiferromagnetic order [7].

The dynamics of the model [Eq. (4)] are simulated following the method in, e.g., Refs. [51–53]. First, an ensemble of configurations is generated at temperature T using a classical Monte Carlo simulation, treating the spins as vectors of fixed length $|\mathbf{S}_i| = 1/2$. We then time evolve the configurations using the Heisenberg equation of motion

$$\hbar\partial_t \mathbf{S}_i(t) = \mathbf{S}_i(t) \times \mathbf{h}_i^{\rm eff}(t), \quad (5)$$

where $\mathbf{h}_i^{\rm eff}(t)$ is the effective exchange field produced by the spins surrounding i. The dynamical structure factor is then calculated by Fourier transforming the correlation functions in both time and space and averaging over the ensemble. We do not include an explicit dissipation term in Eq. (5) but the resultant dynamics can nevertheless be dissipative, due to the strong interactions between modes, arising from nonlinearity.

Since the simulations sample from a classical ensemble of states, the comparison of the phenomenological theory with the simulation results requires using the classical fluctuation-dissipation relationship $S(\omega) = (2k_BT/\omega){\rm Im}[\chi(\omega)]$, as opposed to the quantum relationship $S(\omega) = 2[n_{BE}(\omega)+1]{\rm Im}[\chi(\omega)]$ used to derive Eq. (2) [48]. This leads to the following modified scaling law:

$$\frac{k_B(T-T_{\rm rod})^2}{T} S_{\rm rod}^{\rm cl}(\omega) = \frac{A}{W^2 + (\frac{\hbar\omega}{k_B(T-T_{\rm rod})})^2}; \quad (6)$$

$S_{\rm rod}^{\rm cl}(\omega)$ is the semiclassical structure factor integrated along a $\{111\}$ rod and the right-hand side of Eq. (6) is only a function of the ratio $[\hbar\omega/k_B(T-T_{\rm rod})]$. A and W are nonuniversal constants.

In Fig. 2 we show the scaling collapse of the simulated $S_{\rm rod}^{\rm cl}(\omega)$ for three different sets of exchange parameters $\{J_k\}$. For each parameter set, $T_{\rm rod}$ is treated as an adjustable parameter to optimize the data collapse.

In Fig. 2(a) we show the simulation data for the exchange parameters estimated for $Yb_2Ti_2O_7$ in [32]. This parameter set lies close to the FM-AFM boundary, but not exactly on it. Accordingly, the collapse of the simulation data is close, but imperfect. Adjusting the value of J_1, such that the parameters lie exactly on the $T = 0$ FM-AFM phase boundary, greatly improves quality of the data collapse as shown in Fig. 2(b). Moving away from the phase boundary the collapse becomes worse (see Supplemental

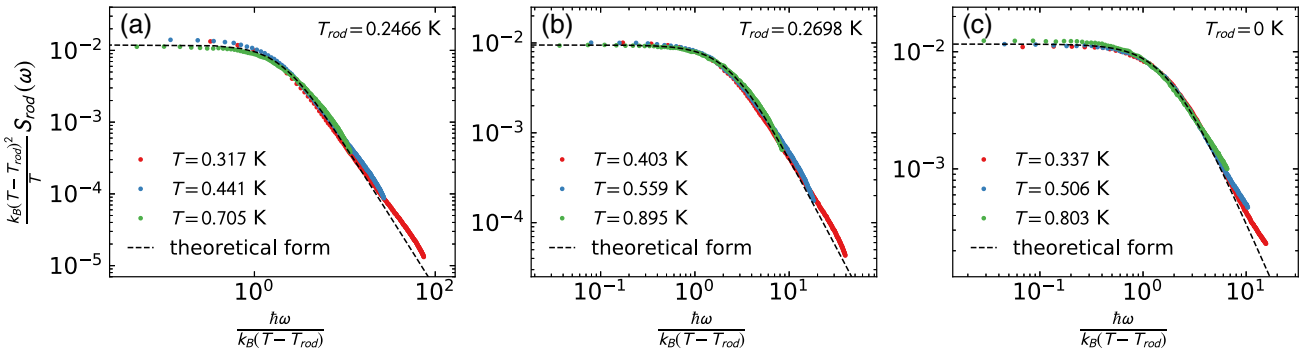

FIG. 2. Dynamical scaling collapse of $S^{\rm cl}_{\rm rod}(\omega)$ calculated using molecular dynamics simulations compared with the theoretical scaling relation Eq. (6). (a) Calculations with exchange parameters set to the values estimated for $Yb_2Ti_2O_7$ in [32] (point A in Fig. 3). A near, but imperfect, collapse is observed. (b) Calculations with a modified value of J_1 such that the exchange parameters lie on the FM-AFM boundary (point B in Fig. 3). A much closer data collapse is observed compared to (a). (c) Calculations at the spin liquid point $J_1 = J_2 = J_4 = 0$, $J_3 < 0$ (point C in Fig. 3). The collapse is observed with a vanishing value of the rod criticality temperature $T_{\rm rod}$.

Material [48]). This confirms the connection between the observed dynamical scaling and the proximity of the FM-AFM phase boundary.

The MD data collapses in Figs. 2(a) and 2(b) both use finite values of $T_{\rm rod}$. In both cases $T_{\rm rod} < T_{\rm order}$, where $T_{\rm order}$ is the temperature of a magnetic ordering transition. Similarly, in experiment $T_{\rm rod} = 0 < T_{\rm order} = 0.27$ K. The point where the rods become critical is thus hidden beneath a thermodynamic phase transition and never reached in the simulations, although its effects are seen in the correlated paramagnetic phase.

A striking aspect of the experimental results is the vanishing value of $T_{\rm rod} \approx 0$, whereas the simulations for parameters close to $Yb_2Ti_2O_7$ find a finite value of $T_{\rm rod}$. The vanishing of $T_{\rm rod}$ is suggestive of the influence of a spin liquid, and indeed there is such a spin liquid on the phase diagram where three ordered phases meet and magnetic order is completely suppressed [7,36]. In Fig. 3 we show how the transition temperatures of FM-AFM phases found in simulation collapse approaching this point, marked C. The temperature scale $T_{\rm rod}$ also tends to zero approaching the spin liquid, as shown in Fig. 4. We

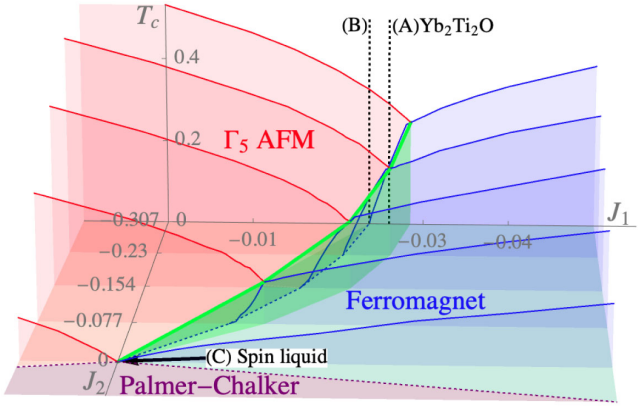

FIG. 3. Finite temperature phase diagram of the pyrochlore $\{J_1, J_2, J_3, J_4\}$ exchange model [17,48,50], determined from classical Monte Carlo simulations. The horizontal axis is J_1, the vertical axis is temperature, and the out-of-the-page axis is J_2, with $J_3 = -0.322$ meV and $J_4 = -0.091 J_2$. The solid lines show $T_{\rm order}$ as a function of J_1, for a series of values of J_2. Point A shows the $Yb_2Ti_2O_7$ exchange parameters [32]. Point B has the same values of $J_{2,3,4}$ as A, but J_1 is adjusted so as to lie exactly on the phase boundary. Point C corresponds to the spin liquid at $J_1 = J_2 = 0$ [36], where FM and Γ_5 orders meet Palmer-Chalker antiferromagnetic order. The green line shows the finite temperature boundary between the ferromagnet (FM) and antiferromagnetic Γ_5 (AFM) states, which goes to zero at the classical spin liquid point. Thus in the finite temperature regime, $Yb_2Ti_2O_7$ is continuously connected to a zero temperature spin liquid phase.

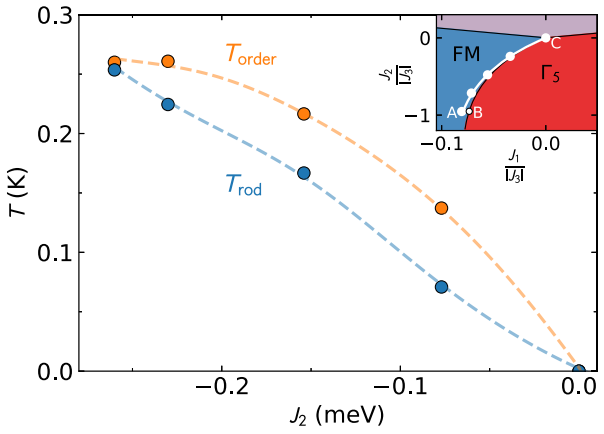

FIG. 4. Variation of thermodynamic transition temperature $T_{\rm order}$, and dynamic criticality temperature $T_{\rm rod}$, found in simulation. Results are shown for a path in parameter space that connects $Yb_2Ti_2O_7$ (A) to the spin-liquid point $J_2 = 0$ (C), shown by a white line in the inset. Both $T_{\rm order}$ and $T_{\rm rod}$ tend to zero approaching the spin liquid. $T_{\rm rod} < T_{\rm ordering}$ for all parameters, meaning that the approach to criticality on the rods is cutoff by the ordering transition as temperature is lowered. The effects of this hidden critical point are nevertheless seen in the paramagnetic phase.

therefore conjecture that the vanishing value of T_{rod} in experiment stems from the influence of a nearby spin liquid, whose regime of influence is widened by quantum fluctuations.

This hypothesis, that the $Yb_2Ti_2O_7$ finite temperature phase is driven by a proximate spin liquid, is reasonable given (i) the finite-temperature regime is continuously connected to the zero-temperature spin liquid, with a smooth decrease of T_{order} connecting the two [Fig. 3], (ii) the observed experimental scaling collapse in $S_{\mathrm{rod}}(\omega)$ with $T_{\mathrm{rod}} = 0$ [Fig. 1(j)] is a feature of the classical spin liquid point [Fig. 2(c)], (iii) spin-wave calculations suggest that quantum fluctuations expand the classical spin liquid to a finite region in parameter space extending especially along the FM-AFM phase boundary [7]. If the continuous connection to a spin liquid phase indicates the presence of unconventional excitations, this may explain the anomalous transport behavior in the $Yb_2Ti_2O_7$ finite-temperature phase [37–40], a topic worth further exploration.

In summary, we have experimentally demonstrated a dynamical scaling relation in the structure factor for inelastic neutron scattering in $Yb_2Ti_2O_7$. We have shown how this scaling can be understood using a phenomenological theory based on multiple phase competition, and demonstrated that equivalent scaling can be found in simulations of a microscopic model of $Yb_2Ti_2O_7$. These results show how multiple phase competition can have universal consequences beyond the ground state, manifesting in the spin dynamics of a correlated paramagnetic phase.

The short-range correlated phase of $Yb_2Ti_2O_7$ is thus best understood in terms of an underlying competition between ferromagnetism and antiferromagnetism and the influence of this competition extends not just to static but also dynamic properties. The description of the dynamics in terms of a Langevin equation suggests that magnetic quasiparticles are either strongly scattered or absent in the paramagnetic regime. Future work will be needed to address whether this theory can explain other mysterious intermediate-temperature behaviors of $Yb_2Ti_2O_7$, such as transport.

Since extended low energy modes are quite a common feature of frustrated magnets in general it seems likely that a similar framework may apply to several materials. In particular, given that a finite-temperature correlated phase is a feature of many Yb^{3+} pyrochlores [54], the phenomenology seen here may prove generic to the entire class, particularly $Yb_2Ge_2O_7$ which also lies close to a phase boundary [55,56]. Moreover, since extended degenerate modes emerge on several phase boundaries of the pyrochlore anisotropic exchange model [Eq. (4)] [7], it would be interesting to search for dynamical scaling behavior in other pyrochlore oxides such as $Er_2Sn_2O_7$ [57–59].

Taking a wider perspective, our experimental results and their interpretation via Eq. (3) imply an emergent relaxation time $\tau_{\mathrm{rod}} = (1/R)(\hbar/k_BT)$ with $R \approx 0.8$ [48]. This is close to the "Planckian" dissipation time $\tau_{\mathrm{Planck}} = (\hbar/k_BT)$ which has been discussed in the context of quantum critical systems as a possible fundamental bound on dissipative timescales [60–63]. Experimental efforts in this area have focussed principally on charge scattering in metals, but if there is a universal principle at play it should presumably show up in other contexts too, including the spin dynamics of correlated insulators. Whether there is any link between these concepts and the physics uncovered here in $Yb_2Ti_2O_7$ is a direction worth exploring.

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