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Understanding Reentrance in Frustrated Magnets: the Case of the $Er_2Sn_2O_7$ Pyrochlore

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Reentrance, the return of a system from an ordered phase to a previously encountered less-ordered one as a controlled parameter is continuously varied, is a recurring theme found in disparate physical systems, yet its microscopic cause is often not investigated thoroughly. Here, through detailed characterization and theoretical modeling, we uncover the microscopic mechanism behind reentrance in the strongly frustrated pyrochlore antiferromagnet $Er_2Sn_2O_7$. We use single crystal heat capacity measurements to expose that $Er_2Sn_2O_7$ exhibits multiple instances of reentrance in its magnetic field B vs. temperature T phase diagram for magnetic fields along three cubic high symmetry directions. Through classical Monte Carlo simulations, mean field theory and classical linear spin-wave expansions, we argue that the origins of the multiple occurrences of reentrance observed in $Er_2Sn_2O_7$ are linked to soft modes. These soft modes arise from phase competition and enhance thermal fluctuations, that entropically stabilize a specific ordered phase, resulting in an increased transition temperature for certain field values and thus the reentrant behavior. Our work represents a detailed examination into the mechanisms responsible for reentrance in a frustrated magnet and may serve as a template for the interpretation of reentrant phenomena in other physical systems.

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Within the field of magnetism, frustration refers to a system's inability to simultaneously satisfy all of its energetic preferences. Strong frustration can result in a variety of exotic phenomena such as spin liquids, spin ice, emergent quasiparticles, topological phases and order-by-disorder [1–7]. Most of the research focus in this area over the past thirty years has been devoted to investigating the physics near zero temperature, considering finite temperatures as a necessary *modus operandi* to search for signatures of the low-energy properties. However, even when subject to high frustration, a majority of frustrated magnetic materials ultimately develop long-range order or display spin-glass freezing at a nonzero critical temperature T_c, albeit often at a very low one compared to the spin-spin interactions. In this context, it therefore seems natural to ask what behavior near T_c may be a witness of the zero-temperature ground state physics. This is particularly important when T_c is just above the experimental baseline temperature, so that temperatures which are low relative to T_c cannot be reached. Here we precisely consider such a situation, as arises in the $Er_2Sn_2O_7$ pyrochlore antiferromagnet, and which provides an opportunity to study a recurrent aspect of frustrated magnetic systems observed at nonzero temperature: reentrance [8–16].

Reentrance occurs when a system, after having developed an ordered phase of some sort, returns to its original less-ordered (e.g. paramagnetic) state as some parameter (e.g. temperature, field, pressure, stoichiometry) is continuously varied. Reentrance has been found in spin glasses [17, 18], liquid mixtures [19, 20], protein thermodynamics [21], liquid crystals [22, 23], bilayer graphene [24], superconductors [25], modulated phases [26, 27] and even in black hole thermodynamics [28]. Despite its ubiquity, reentrance is typically unexpected and its explanation in terms of entropic contributions to the free-energy from the underlying microscopic degrees of freedom is usually subtle. In this context, while frequently observed in frustrated magnets, the microscopic mechanism leading to reentrance often remains obscure [8–15]. Two mechanisms have commonly been invoked: a field-dependent suppression of quantum fluctuations [29–31] and the partial disorder of an intervening phase [18, 32–34]. Here, we present an alternative scenario of a generic nature which illustrates how the observation of reentrance may be used as a fingerprint of the frustration at play in the ground state.

In this article, we show that $Er_2Sn_2O_7$ represents a tractable material example where the intricate microscopic mechanisms responsible for reentrance in frustrated magnets can be rigorously studied experimentally and theoretically. $Er_2Sn_2O_7$ holds a special place among rare earth pyrochlores [5, 35]: it is well-characterized, has a suppressed critical temperature and is one of the few materials with a simple Palmer-Chalker (PC) antiferromagnetic ground state [36–38] [Fig. 1(a)]. Its estimated exchange and single-ion susceptibility parameters are highly anisotropic and theory suggests a proximity to another competing antiferromagnetic phase [37–39] known as Γ_5 [5, 6, 39] [Fig. 1(b,c)]. Because of this anisotropy, the

response of $Er_2Sn_2O_7$ to an applied field is expected to differ with field direction, as has been poignantly illustrated with the experimental exploration of rare earth pyrochlore titanates [5, 6, 11, 12, 35]. As such, the present study critically relies on the recently gained, and notoriously challenging, ability to synthesize pyrochlore stannate single crystals [40], including $Er_2Sn_2O_7$.

We report herein the discovery of five occurrences of reentrance in the B-T phase diagram of $Er_2Sn_2O_7$ for fields along the [100], [110] and [111] cubic directions using heat capacity measurements. By thoroughly investigating this experimental phase diagram using mean field theory, classical linear spin-wave expansions and Monte Carlo simulations, we have uncovered the various, and distinct, microscopic origins of reentrance in this system. In short, we find that multi-phase competitions at $T = 0$ result in enhanced thermal fluctuations at specific field values. These fluctuations *entropically* stabilize the corresponding *ordered* phase over the disordered phase, and thus increase $T_c(B)$ over a certain B field range. This produces $T_c(B)$ reentrant phase boundaries whose maximal temperature extent corresponds approximately to the zero-temperature field-driven phase transitions [see Fig. 2(a-c)]. This multi-phase competition is in some cases a direct consequence of the competition of the field-evolved PC states while in others it is inherent to $Er_2Sn_2O_7$'s zero-field ground state being in close proximity to the phase boundary between the PC and Γ_5 phases. See the Supplementary Material [45] for technical details on the experiments, simulations and analytics.

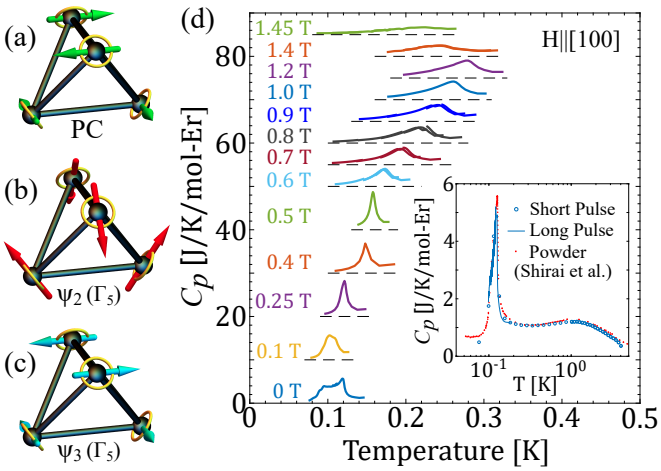

FIG. 1: Example of 6-fold degenerate states: (a) Palmer-Chalker [36] and (b) ψ_2 and (c) ψ_3 basis states of Γ_5 [46]. The ψ_2 and ψ_3 states are connected by a rotation of the spins by an angle ϕ within their local easy-planes (yellow circles): $\phi \equiv n\pi/3$ $(+\pi/6)$ for $n = 0, ..., 5$ correspond to ψ_2 (ψ_3) [47]. Panels (b) and (c) are for $\phi = 0$ and $\pi/2$, respectively. The manifold with U(1) degeneracy, $\phi \in [0, 2\pi]$, forms the so-called Γ_5 states that appear in the [111] phase diagram. (d) Heat capacity, $C_p(T)$, vs temperature, T, of $Er_2Sn_2O_7$ with the magnetic field along [100], showing the reentrant nature of the transition. Curves at different fields are offset vertically for clarity. Similar data for the [110] and [111] field directions are included in the Supplementary Material [45]. (Inset) $C_p(T)$ $Er_2Sn_2O_7$ in zero-field, with short and long pulse measurements on crystal samples overlaid. Powder data from Shirai *et al.* [48] is also overlaid to demonstrate agreement between sample types.

Heat capacity (C_p) measurements were performed on single crystal samples, grown via the hydrothermal method described in Ref. [40], down to 50 mK with varying magnetic fields, $\boldsymbol{B}$, oriented in the [111], [110], and [100] directions, using a dilution refrigerator insert in a Quantum Design Physical Properties Measurement System. Two measurement techniques were used: the conventional quasi-adiabatic thermal relaxation method (called "short pulses" hereafter), as well as "long pulses", both of which are described in detail in Ref. [49]. The long pulse technique allows faster and higher point-density measurements across phase transitions, enabling an accurate mapping of a phase diagram by measuring the field dependence of the transition temperature, $T_c(B)$ [Fig. 1 (d)].

In the zero-field heat capacity [Fig. 1 (d) inset], we find a sharp lambda-like anomaly indicative of a phase transition at $T_c = 118 \pm 5$ mK, which is consistent with previous measurements on powder samples reported in Ref. [48] (130 mK, from heat capacity, data shown in Fig. 1 (d) inset) and Ref. [38] (108 ± 5 mK, from DC magnetic susceptibility). The extremely high point density of the long pulse measurements allow for the observation of subtle features in the peak shape, which are typically not resolved by conventional short pulse measurements. This reveals a low temperature shoulder of the C_p peak in the zero-field data at 97 ± 5 mK [45]. We performed elastic neutron scattering measurements to determine the magnetic structure between the sharp high temperature peak and the low temperature shoulder to check for an intermediate magnetic phase [45]. We found that the magnetic structure is of Palmer-Chalker type at all measured temperatures throughout the transition range with no sign of other magnetic phases. It is not clear what causes this structure in the heat capacity anomaly, but we note that similar (though not identical) broadening is observed in all five crystals we have measured as well as in published data on a powder sample [45, 48] (Fig. 1 (d) inset). Thus, it seems to be a feature of all $Er_2Sn_2O_7$ samples, but is likely due to (or influenced by) slight inhomogeneities rather than being purely intrinsic in origin. Although it may be worth future investigation, its presence does not affect any of the conclusions of this work.

To model $Er_2Sn_2O_7$, we use the generic nearest-neighbor Hamiltonian on the pyrochlore lattice [39, 50],

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta - \mu_B \sum_i g_i^{\alpha\beta} B^\alpha S_i^\beta. \quad (1)$$

$\boldsymbol{S}_i = (S_i^x, S_i^y, S_i^z)$ is a three-component pseudo-spin of length $|\boldsymbol{S}_i| = 1/2$ and $\boldsymbol{B}$ is the external magnetic field. The g-tensor represents the single-ion anisotropy, with local easy-plane $g_\perp$ and easy-axis $g_\parallel$ components at lattice site i. Given the symmetries of the pyrochlore lattice, the anisotropic exchange matrix $J_{ij}^{\alpha\beta}$ is parameterized by four independent cou-

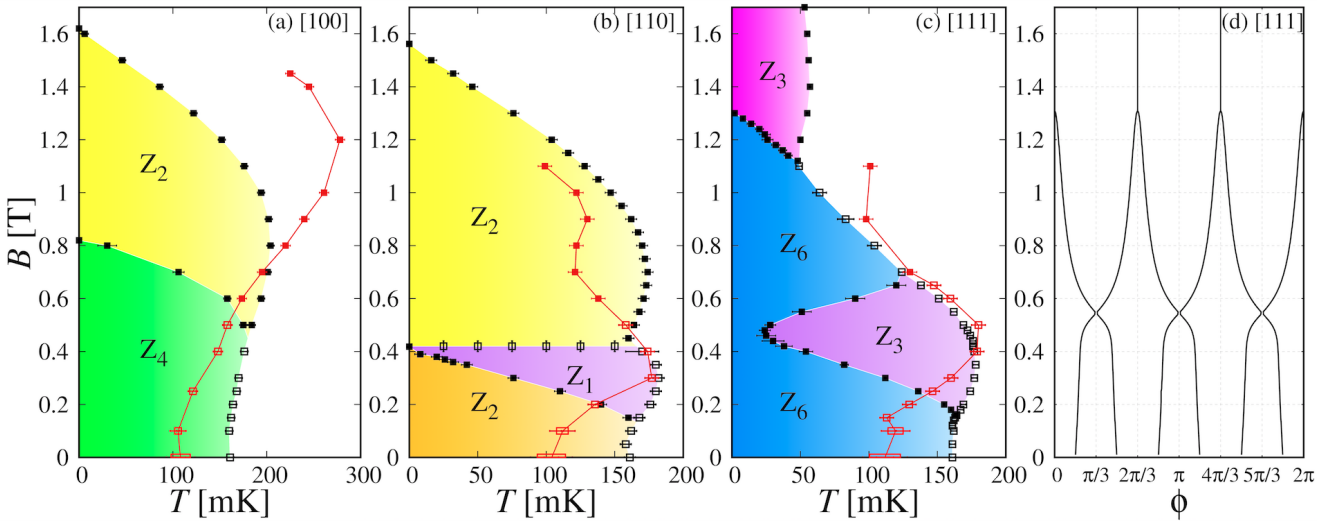

FIG. 2: B-T phase diagrams of $Er_2Sn_2O_7$ in the (a) [100], (b) [110] and (c) [111] field directions, comparing experimental data with sharp (□) and smooth (■) heat-capacity peaks [Fig. 1(d)], to Monte Carlo results with 1st (□) and 2nd (■) order transitions. Experiments and simulations are notably similar, showing the same (multiple) reentrance. The degeneracy Z_n found in simulations is given for each phase. The width of the red rectangles at 0 and 0.1 T represent the position of the double peaks. (d) In a [111] field, each of the six FEPC ground states has a Γ_5 contribution described by an angle ϕ [Fig. 1(b,c)], that can be computed exactly by minimizing the energy of one tetrahedron as a function of B.

pling constants: (J_1, J_2, J_3, J_4) [50, 51]. $Er_2Sn_2O_7$ has been previously parameterized using inelastic neutron scattering on powder samples [37, 38]. Here we choose to remain within the error bars of Ref. [38], selecting a set of coupling parameters where simulations find $T_c \sim 180$ mK at 0.4 T for a [111] field to match the experimental result [61]: $(J_1, J_2, J_3, J_4) = (+0.079, +0.066, -0.111, +0.032)$ meV and $g_\perp = 7.52, g_\parallel = 0.054$. Note that the nearest-neighbor part of dipolar interactions is included in the $J_{ij}^{\alpha\beta}$ couplings of the Hamiltonian (1).

To proceed, we first analyze this model using classical Monte Carlo simulations, with the results summarized in the B-T phase diagrams of Fig. 2. Most importantly, with $T_c(B = 0.4\,\mathrm{T})$ fitted (for $\boldsymbol{B}$ along the [111] direction), the simulations reproduce the number of reentrant "lobes" for each field direction (e.g. one and two for a [100] and [110] field, respectively), as well as, at each lobe, the rough magnitude of the increase of T_c at the corresponding value of B. Moreover, simulations find that the transition always evolves from discontinuous to continuous when increasing the field. This is consistent with the shape of the experimental heat capacity peaks, evolving from sharp to smooth, and a further hint that simulations are capturing the proper physics displayed by the experiments. We suspect that a fine-tuning of the $J_{ij}^{\alpha\beta}$ coupling parameters and incorporating quantum fluctuations, as well as perhaps dipolar interactions beyond nearest neighbors, should account for the quantitative disagreements. Nevertheless, the semi-quantitative match between experiments and simulations confirms the validity of Eq. (1) as a minimal model for $Er_2Sn_2O_7$, suggesting that simulations robustly encapsulate the key physics behind the experimentally observed multiple occurrences of reentrance.

The results in Figs. 2(a-c) raise multiple questions. Why are there multiple instances of reentrance and why are they so strongly dependent on the field direction? More fundamentally, why does $Er_2Sn_2O_7$ demonstrate reentrance in the first place? As a set of clues, simulations bring to light a variety of phases that vie for ordering. In the rest of this article, we explain how soft modes induced by this multi-phase competition are linked to reentrance, using a combination of mean field theory and classical linear spin-wave expansions.

The zero-field ground state of $Er_2Sn_2O_7$ is the sixfold-degenerate PC phase. However, the ground states naturally deform and evolve under the application of a magnetic field. For sufficiently large fields, some of these field-evolved PC (FEPC) states may become partially polarized into the *same* spin configuration. We therefore label the resulting phase according to the number of FEPC states that minimize the free energy but have *distinct* spin configurations (e.g. Z_6 at $B = T = 0$, for the six degenerate PC states, and Z_1 at sufficiently large B for the trivial field-polarized paramagnet). Phase transitions then occur whenever distinct FEPC states "merge" into the same spin configuration at a given field value B_c.

First, consider the [100] field phase diagram in Fig. 2(a). At $T = 0$, the FEPC states merge at $B_c = 0.82$ T, giving rise to the yellow Z_2 region. Fig. 3 displays the classical spin-wave dispersions $\kappa_\nu(\boldsymbol{q})$ for a number of field values below and above B_c, calculated from the corresponding $T = 0$ FEPC ground states. As the merger transition is approached at $B_c = 0.82$ T, the bottom of the dispersive bands drop below the energy scale set by $T_c(B = 0) \approx 160$ mK, becoming soft and gapless at $B = B_c$. This decrease indicates a propensity for stronger thermal fluctuations at B_c than at other field values. More precisely, since $s = -\frac{1}{8N_q}\sum_{\boldsymbol{q}}\sum_{\nu=1}^{8}\ln(\kappa_\nu(\boldsymbol{q}))$ quantifies the

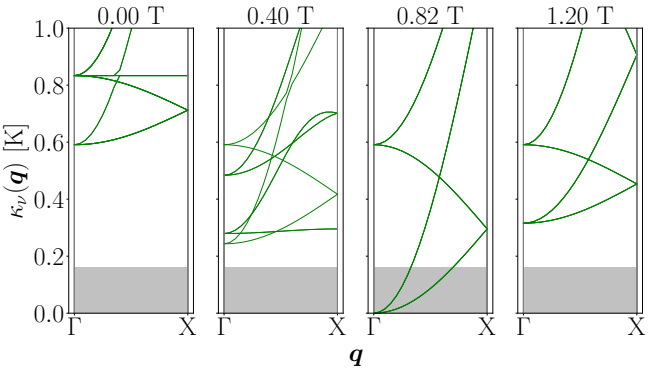

FIG. 3: Classical spin-wave dispersions for $B = 0, 0.40, 0.82$, and 1.20 T along the [100] direction, for a path in the FCC Brillouin zone. Note that $B_c = 0.82$ T is a critical field at $T = 0$, as shown in Fig. 2. The grey boxes indicate energy scales below $T_c(B = 0\,\mathrm{T}) \approx 160$ mK from Monte Carlo simulations; when modes occur within this region they are considered "soft". Note that the dispersions for *all* Palmer-Chalker states are plotted, but may overlap at high-symmetry points or due to their degeneracies in a field.

entropy contribution from classical spin-waves, the decrease in $\kappa_\nu(\boldsymbol{q})$ on approaching B_c from above or below (as shown in Fig. 3) corresponds to an *increase* in entropy within the ordered phase. As a consequence, the gapless soft modes at B_c stabilize the yellow Z_2 region of Fig. 2 at finite temperature, both over the green Z_4 region as well as the disordered paramagnet.

At first sight, the above discussion may remind the reader of order-by-disorder, but the two mechanisms are in general different. Specifically, the Z_2 selection for $B \geq B_c$ is energetic and *not* entropic. As opposed to order-by-disorder, soft modes do not select the Z_2 states among a degenerate manifold, but rather enhance their entropic stability at a specific field, B_c, compared to higher and lower fields. This pushes the transition temperature upwards around B_c, inducing reentrance.

The microscopic physics at play is different in a [111] field; simulations reveal a reentrant lobe around a field value (~ 0.4 T) for which no corresponding $T = 0$ FEPC merger is found in the calculations. To understand this reentrance, it is important to note that the long-range order of $Er_2Sn_2O_7$ in a [111] field is not described by a single irreducible representation (irrep) [39]. Instead, it is described by the naturally field-induced ferromagnetic irrep as well as the Γ_5 irrep [Fig. 1(b,c)] due to the proximity of the Γ_5 ground state to the PC phase in zero-field [39]. The Γ_5 states bear an accidental U(1) degeneracy parameterized by an angle ϕ [39, 47, 52–54], which is lifted by a magnetic field [55] with discrete values of ϕ being selected, as shown in Fig. 2(d). While the six FEPC states remain distinct in this region, their Γ_5 components merge at $B \approx 0.55$ T into three $\phi = \{\pi/3, \pi, 5\pi/3\}$ corresponding to three of the six ψ_2 states [Fig. 1(b), Ref. [45]]. This ψ_2 selection is associated with a flat low-energy soft mode in the spin-wave expansion at $B \approx 0.5$ T and simulations confirm the presence of partial ψ_2 order in the reentrant lobe, shown by the violet Z_3 phase in Fig. 2(c). These results make a strong case unraveling the mechanism of reentrance; the intervening Z_3 phase,which is *n*ot part of the ground states, is entropically stabilized by low-energy soft modes arising from the PC and $\Gamma_5(\psi_2)$ phase competition.

Closing the [111] case, one should mention the FEPC merger transition at $B_c = 1.31$ T and $T = 0$ is naturally accompanied by a merging of the ϕ values (here also corresponding to ψ_2 states, see Fig. 2(d)) and by a small reentrant lobe [Fig. 2(c)], as expected from the discussion for the [100] field case. Our experimental data point towards the onset of this high-field lobe as well (see Fig. 2(c) for $B = 0.9$ T & 1.1 T). However, it was not possible to explore this high-field region experimentally because the sample did not easily equilibrate above 0.7 T. Interestingly, simulations also suffer from difficulties thermalizing between 0.7 and 1.2 T.

Finally, the mechanisms behind reentrance for a [110] field are reminiscent of the other two field directions [Fig. 2(b)]. Below $B \lesssim 0.1$ T, simulations are difficult to thermalize [45], but above $B \gtrsim 0.1$ T, we find two FEPC ground states that merge at $B_c = 0.42$ T. This merging gives rise to gapless soft modes, the subsequent violet Z_1 phase, and reentrance at finite temperatures. It is the same mechanism as in a [100] field. However, as opposed to the [100] scenario, this newly merged ground state vanishes immediately once $B > B_c$ (i.e. it becomes an excited state). The system is then found in two *other* ground states (corresponding to the yellow Z_2 phase). The vanishing of the merged state corresponds to the Z_1 phase abruptly disappearing above B_c and the removal of the aforementioned gapless soft modes. This causes the rapid collapse of the reentrant lobe at $B \sim 0.42$ T. The fact that Z_1 order is only stable at one point at $T = 0$ is reminiscent of the triangular Heisenberg antiferromagnet in a field, where an intervening $1/3$ plateau also spreads at finite temperature and gives rise to reentrance [8, 13, 56] – a mechanism that might be at play in $Ba_3CoSb_2O_9$ [57]. However, the reentrance phenomenon in our system at 0.42 T differs from this case in that it does not require the extensive degeneracy of a magnetization plateau, but simply the more generic presence of soft modes about a long-range ordered non-degenerate spin configuration.

At higher [110] field, another reentrant dome appears ($B \sim 0.7$ T), however, there are no ground state FEPC mergers involved. Similarly to the reentrant behavior at low [111] field, spin wave theory shows a *minimum (non-zero) gap* that entropically favors the ordered phase around 0.7 T (see Section S5 in Ref [45]). These low-energy gapped modes, along with the collapse at $B_c = 0.42$ T, give the higher field reentrant lobe at ~ 0.7 T.

In summary, we have presented the first exploration of the field-direction dependence of the thermodynamics of stannate pyrochlores, which, despite decades of effort, were not available as single crystals until very recently [40]. Access to these crystals has proven to be crucial since the phase diagram of $Er_2Sn_2O_7$ is highly sensitive to the field direction, and exhibits several reentrant lobes with sundry underlying mechanisms. These features result from the competition of several orders, especially the zero-field Palmer-Chalker, the field-induced fer-

romagnetic and the neighboring Γ_5 states. In particular, most instances of reentrance in the phase diagram can be traced to zero-temperature field-induced merging of ground states. This energetic selection is accompanied by soft modes which entropically enhance the transition temperatures, and this mechanism is thus distinct from the one of order-by-disorder. In this light, reentrance is a useful and experimentally accessible fingerprint at the critical temperature of an underlying zero-temperature phase transition.

Given that multi-phase competition is a common feature of frustrated magnetism, we expect the mechanisms we have uncovered to be widespread among magnetic systems displaying reentrance; especially since it does not require the accidental presence of an exotic partially-disordered phase [18, 33, 58–60] or a phase with extensive entropy, such as in a magnetization plateau state [8, 13, 56, 57]. In semi-classical and quantum systems, our mechanism may work together with the field-induced suppression of quantum fluctuations [29] to produce even larger reentrant lobes. We hope our work will motivate others to pursue a microscopic interpretation of future observations of reentrance (and possibly to revisit old ones [8–16]) in light of zero-temperature transitions. Since magnetic systems often afford us with minimal models to understand other areas of physics, our results raise a more general question: if reentrance is observed by varying a given parameter, when is it actually due to a nearby transition in a broader, and perhaps not even physically accessible, parameter space?

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