Pseudo-Goldstone Gaps and Order-by-Quantum Disorder in Frustrated Magnets

Jeffrey G. Rau, Paul A. McClarty, and Roderich Moessner
Max-Planck-Institut für Physik komplexer Systeme, 01187 Dresden, Germany

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In systems with competing interactions, continuous degeneracies can appear which are accidental, in that they are not related to any symmetry of the Hamiltonian. Accordingly, the pseudo-Goldstone modes associated with these degeneracies are also unprotected. Indeed, through a process known as "order-by-quantum disorder," quantum zero-point fluctuations can lift the degeneracy and induce a gap for these modes. We show that this gap can be exactly computed at leading order in $1/S$ in spin-wave theory from the mean curvature of the classical and quantum zero-point energies—without the need to consider any spin-wave interactions. We confirm this equivalence through direct calculations of the spin-wave spectrum to $O(1/S^2)$ in a wide variety of theoretically and experimentally relevant quantum spin models. We prove this equivalence through the use of an exact sum rule that provides the required mixing of different orders of $1/S$. Finally, we discuss some implications for several leading order-by-quantum-disorder candidate materials, clarifying the expected pseudo-Goldstone gap sizes in $Er_2Ti_2O_7$ and $Ca_3Fe_2Ge_3O_{12}$.

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Goldstone's theorem [1] connects the spontaneous breaking of a continuous symmetry to the presence of gapless excitations—a foundational result with applications in almost every branch of physics. Alternatively, gapless excitations can be generated by *accidental* degeneracies which are not symmetry enforced [2]. Just as continuous symmetries imply the presence of gapless Nambu-Goldstone modes, accidental degeneracies imply the presence of pseudo-Goldstone modes that are *nearly* gapless when (inevitably) these degeneracies are weakly lifted. Such modes have been invoked to explain the appearance of unexpectedly low-lying excitations many contexts, ranging from quantum chromodynamics [3] to high-temperature superconductors [4,5] and quantum magnets [6–8]; perhaps the most well known example is the mass of the pion, which arises due to broken chiral symmetry [9,10].

In lieu of explicit symmetry breaking, accidental degeneracies can also be lifted by *fluctuations*. Broadly referred to as "order by disorder" [6–8], this phenomenon has proven useful in understanding a wide variety of ordering phenomena in frustrated spin systems [7,11–14], where accidental degeneracies are natural. An example is "order-by-quantum disorder" [7,8], where an accidentally degenerate manifold in the classical limit, $S \to \infty$, is lifted by quantum corrections at $O(1/S)$. Within noninteracting spin-wave theory [15], these contributions can be viewed as the zero-point energy of the harmonic spin-waves selecting some subset of the classically degenerate manifold [7,8].

Through order-by-quantum disorder, the pseudo-Goldstone modes associated with this accidental degeneracy must acquire a gap. Since in noninteracting spin-wave theory these modes are gapless, to obtain a finite gap one must include the effects of spin-wave interactions. While conceptually simple, such calculations have only been carried out for a few limited cases, mostly for simple isotropic Heisenberg-like models [8,14,16–20]. In more complex models with strong exchange anisotropy, such as in the rare-earth pyrochlores [21] or in Kitaev magnets [22,23], these calculations can be complicated by the presence of three-magnon interactions that can lead to spontaneous magnon decay [24], even for colinear magnetic ground states.

In this Letter, we offer a significant simplification, showing that the curvatures of classical and quantum zero-point energy densities computed at $O(1/S)$, are already sufficient to determine the pseudo-Goldstone gap exactly to $O(1/S^2)$. Explicitly, if the classically degenerate manifold is parametrized by ϕ with conjugate direction θ (see Fig. 1), the pseudo-Goldstone gap, Δ, is given by [25]

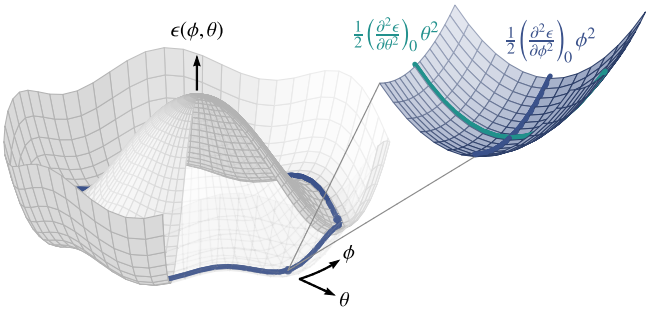

FIG. 1. Schematic illustration of the semiclassical energy density $\epsilon(\phi,\theta)$, and the curvatures $[(\partial^2\epsilon/\partial\theta^2)]_0$ and $[(\partial^2\epsilon/\partial\phi^2)]_0$ about the semiclassical ground state, which are related to the pseudo-Goldstone gap, Δ, via Eq. (1). The nearly soft manifold associated with a type I pseudo-Goldstone mode is shown, with the two principal curvatures about a global minimum indicated.

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$$\Delta = \frac{1}{S}\sqrt{\left(\frac{\partial^2\epsilon}{\partial\theta^2}\right)_0\left(\frac{\partial^2\epsilon}{\partial\phi^2}\right)_0 - \left(\frac{\partial^2\epsilon}{\partial\theta\partial\phi}\right)_0^2}, \qquad (1)$$

where the semiclassical energy density, $\epsilon(\phi,\theta)$, of the classical ground state at (ϕ,θ) includes the classical $[O(S^2)]$ and quantum zero-point $[O(S)]$ contributions. While used as a heuristic in several works [8,16,59–61], its equivalence to the leading result from nonlinear spin-wave theory is far from evident in perturbation theory, as it involves mixing of different orders in $1/S$. This formula eliminates much of the burden of computing the pseudo-Goldstone gap, requiring only quantities from standard noninteracting spin-wave theory, a computation considerably more straightforward to undertake in practice.

In light of this, we revisit a variety of models that exhibit order-by-quantum disorder, including square and cubic Heisenberg-compass models [16], Heisenberg-Kitaev-Γ models [60,62] on the honeycomb lattice, and $J_1 - J_2$ models on the square and triangular lattices [7,13,14,63]. For each, we compute the gap both explicitly in interacting spin-wave theory—often for the first time—and then again using the curvature formula [Eq. (1)], confirming that they are indeed identical.

Finally, we consider applications; while order-by-quantum disorder has a long theoretical history [6–8,16,64], there are only a handful of serious potential experimental candidates [8,17,61,65–67]. Two of the best material examples are the cubic Heisenberg antiferromagnet $Ca_3Fe_2Ge_3O_{12}$ [8,65], and the pyrochlore XY antiferromagnet $Er_2Ti_2O_7$ [61,67,68], where the leading energetic effects [69] are naïvely [70–72] expected to be small [61]. In this context, the pseudo-Goldstone gap provides a quantitative benchmark which may be used to distinguish order-by-quantum disorder from more conventional energetic selection. Given knowledge of the models for these materials [61,65] and when the semiclassical picture is a good description, our result provides a straightforward way to estimate the pseudo-Goldstone gap observed experimentally, cleanly demonstrating the utility of these results.

Spin-wave theory.—We first review the physics of pseudo-Goldstone modes as they appear in linear spin-wave theory. As for the usual Goldstone modes, these can be classified into two types [73,74], which we denote as I and II, which correspond to having nonconserved and conserved order parameters, respectively. For a type I pseudo-Goldstone mode the linear spin-wave dispersion vanishes linearly $\sim|\boldsymbol{k}|$, while for the type II case it vanishes quadratically $\sim|\boldsymbol{k}|^2$, as illustrated in Fig. 2.

More explicitly, we can define the linear spin-wave Hamiltonian [15]

$$S\sum_{\mathbf{k}}\sum_{\alpha\beta}\left[A_{\boldsymbol{k}}^{\alpha\beta}a_{\boldsymbol{k}\alpha}^\dagger a_{\boldsymbol{k}\beta} + \frac{1}{2}(B_{\boldsymbol{k}}^{\alpha\beta}a_{\boldsymbol{k}\alpha}^\dagger a_{-\boldsymbol{k}\beta}^\dagger + \text{H.c.})\right], \qquad (2)$$

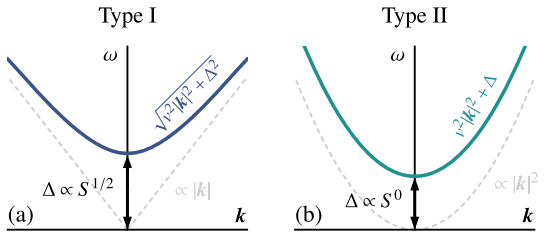

FIG. 2. Schematic form of the spectrum of (a) type I and (b) type II pseudo-Goldstone modes. Type I modes have $\omega \sim |\boldsymbol{k}|$ at $O(S)$, with the pseudo-Goldstone gap scaling as $\Delta \sim O(S^{1/2})$, while type II modes have $\omega \sim |\boldsymbol{k}|^2$ at $O(S)$, with $\Delta \sim O(S^0)$.

where $a_{\boldsymbol{k}\alpha}$ is the (Fourier-transformed) Holstein-Primakoff boson with wave vector $\boldsymbol{k}$ on sublattice α of the (magnetic) unit cell. The matrices $\boldsymbol{A}_{\boldsymbol{k}}$ and $\boldsymbol{B}_{\boldsymbol{k}}$ depend on the classical ordering pattern and the exchange model; see Supplemental Material [25] for details. The linear spin-wave spectrum is determined by the eigenvalues of the Bogoliubov dispersion matrix [75]

$$\boldsymbol{\sigma}_3\boldsymbol{M}_{\boldsymbol{k}} \equiv \begin{pmatrix} \boldsymbol{A}_{\boldsymbol{k}} & \boldsymbol{B}_{\boldsymbol{k}} \\ -\boldsymbol{B}_{\boldsymbol{k}}^\dagger & -\boldsymbol{A}_{-\boldsymbol{k}}^\intercal \end{pmatrix}, \qquad (3)$$

where $\boldsymbol{\sigma}_3 \equiv \mathrm{diag}(+\mathbf{1}, -\mathbf{1})$ is a block Pauli matrix. A pseudo-Goldstone mode appears as a zero in the linear spin-wave spectrum. Without loss of generality, we assume that this zero mode lies at the zone center, with $\boldsymbol{M}_0$ being positive semidefinite, and $\boldsymbol{M}_{\boldsymbol{k}}$ positive definite elsewhere (this is always possible for commensurate magnetic orders).

When spin-wave interactions are included, the excitation energies are indicated by the poles of the (retarded) magnon Green's function

$$\boldsymbol{G}^R(\boldsymbol{k},\omega) \equiv [(\omega + i0^+)\boldsymbol{\sigma}_3 - S\boldsymbol{M}_{\boldsymbol{k}} - \boldsymbol{\Sigma}^R(\boldsymbol{k},\omega)]^{-1}, \qquad (4)$$

where $\boldsymbol{\Sigma}^R(\boldsymbol{k},\omega)$ is the (retarded) self-energy. We use a formalism where the free magnon Green's function is defined as a matrix that includes both the sublattice indices and the normal and anomalous contributions [75]. The effects of spin-wave interactions encoded in the self-energy can be computed perturbatively [76] in the limit $1/S \to 0$ [77,78]. The leading contributions at order $O(S^0)$ are illustrated in Fig. 3.

The determination of the poles of the magnon Green's function then proceeds perturbatively in the self-energy, with respect to $\boldsymbol{M}_{\boldsymbol{k}}$. For the type I case, one finds the relevant low-energy subspace of $\boldsymbol{\sigma}_3\boldsymbol{M}_0$ is similar to a defective Jordan block [75]. The pseudo-Goldstone gap, Δ, at leading order is then

$$\Delta = S^{1/2}\sqrt{2\mathrm{Re}[\boldsymbol{V}_0^\dagger\boldsymbol{\Sigma}^R(\mathbf{0},0)\boldsymbol{\sigma}_3\boldsymbol{M}_0\boldsymbol{V}_0]} + O(S^{-1/2}), \qquad (5)$$

FIG. 3. The three classes of (Hugenholtz) diagrams contributing to the $O(S^0)$ magnon self-energy, $\boldsymbol{\Sigma}^R(\boldsymbol{k},\omega)$. The (free) propagators (dark line) include normal and anomalous parts, and are connected to external legs (dull line). The first diagram involves a single four-magnon interaction (filled rectangle), while the second and third involve a pair of three-magnon interactions (filled circle).

where $\boldsymbol{V}_0$ characterizes part of the zero mode subspace [25]. For the type II case, there are two linearly independent eigenvectors of $\boldsymbol{\sigma}_3\boldsymbol{M}_0$ with eigenvalue zero, $\boldsymbol{V}_0$ and $\boldsymbol{W}_0$. The pseudo-Goldstone gap is then

$$\Delta = \sqrt{(\boldsymbol{V}_0^\dagger \boldsymbol{\Sigma}^R(\boldsymbol{0},0)\boldsymbol{V}_0)^2 - |\boldsymbol{V}_0^\dagger \boldsymbol{\Sigma}^R(\boldsymbol{0},0)\boldsymbol{W}_0|^2} + O(S^{-1}), \quad (6)$$

at leading order. Once the self-energy due to magnon-magnon interactions is computed, these equations [Eqs. (5) and (6)] allow the direct calculation of the pseudo-Goldstone gap. A detailed derivation for both cases is provided in the Supplemental Material [25].

These follow distinct scalings with the spin length [see Eqs. (5) and (6)]: for a type I pseudo-Goldstone mode the gap scales as $\Delta \sim O(S^{1/2})$, while for a type II pseudo-Goldstone mode it scales as $\Delta \sim O(S^0)$. Away from the zone center [74], the spectrum takes the low-energy form $\sim\sqrt{v^2|\boldsymbol{k}|^2 + \Delta^2}$ for type I modes, while for type II modes it takes the form $\sim v^2|\boldsymbol{k}|^2 + \Delta$, as shown schematically in Fig. 2.

Curvature formula and semiclassical dynamics.—We now motivate the curvature formula for the pseudo-Goldstone gap, Δ [Eq. (1)], through a heuristic semiclassical argument. It is useful to construct a local frame $(\hat{\boldsymbol{x}}_\alpha, \hat{\boldsymbol{y}}_\alpha, \hat{\boldsymbol{z}}_\alpha)$ where $\hat{\boldsymbol{z}}_\alpha$ is the ordering direction, $\hat{\boldsymbol{x}}_\alpha$ is the soft-mode direction, and $\hat{\boldsymbol{y}}_\alpha = \hat{\boldsymbol{z}}_\alpha \times \hat{\boldsymbol{x}}_\alpha$. If we parametrize the soft mode by an angle ϕ, and the (locally) orthogonal directions by an angle θ, we can define the classical spin configuration

$$\boldsymbol{S}_\alpha = S(\phi\hat{\boldsymbol{x}}_\alpha + \theta\hat{\boldsymbol{y}}_\alpha + \hat{\boldsymbol{z}}_\alpha[1 - (\phi^2 + \theta^2)]^{1/2}), \quad (7)$$

accurate to quadratic order in θ and ϕ. For simplicity, we have assumed here that the soft mode is uniform, with the relative weight of the rotations not varying between sublattices—this assumption is not essential, and can be lifted [25]. These variables have the Poisson bracket $\{\phi,\theta\} = NS$ and thus essentially behave like a position and its canonically conjugate momentum. For the type I case, ϕ is classically soft, with no restoring force, while θ is not, while for the type II case both are classically soft.

If we treat these collective coordinates as classical dynamical variables, then quantum fluctuations can be included in an *ad hoc* way by using the semiclassical spin-wave energy density $\epsilon(\phi,\theta)$ as an effective potential; explicitly,

$$\epsilon(\phi,\theta) \equiv \begin{cases} S^2\epsilon_{\rm cl}(\theta) + S\epsilon_{\rm qu}(\phi,0), & \text{type I} \\ S\epsilon_{\rm qu}(\phi,\theta), & \text{type II} \end{cases}, \quad (8)$$

where $\epsilon_{\rm cl}$ is the classical energy density and $\epsilon_{\rm qu}$ is the quantum zero-point energy density [25] computed in linear spin wave theory for the soft spin configurations [Eq. (7)]. Since the θ direction is not soft for type I pseudo-Goldstone modes, the classical part of the energy must be included. The quantum zero-point energy density is defined as $\epsilon_{\rm qu}(\phi,\theta) \equiv (2N)^{-1}\sum_{k\alpha}\epsilon_{k\alpha}(\phi,\theta)$, where $\epsilon_{k\alpha}(\phi,\theta)$ are the spin-wave energies found expanding about a classical ground state with finite ϕ,θ. For the type II case, this zero-point energy is well defined for arbitrary ϕ and θ, with the classical energy independent of both variables; while for the type I case, the zero-point energy is ill defined for $\theta \neq 0$, and thus we fix $\theta = 0$.

The curvatures of the total semiclassical energy density directly determine the normal mode frequency of θ and ϕ via the classical equations of motion [79]

$$\frac{d\phi}{dt} = +\frac{1}{S}\frac{\partial\epsilon}{\partial\theta} \approx +\left(\frac{\partial^2\epsilon}{\partial\theta\partial\phi}\right)_0\phi + \left(\frac{\partial^2\epsilon}{\partial\theta^2}\right)_0\theta, \quad (9a)$$

$$\frac{d\theta}{dt} = -\frac{1}{S}\frac{\partial\epsilon}{\partial\phi} \approx -\left(\frac{\partial^2\epsilon}{\partial\phi^2}\right)_0\phi - \left(\frac{\partial^2\epsilon}{\partial\theta\partial\phi}\right)_0\theta, \quad (9b)$$

giving the pseudo-Goldstone gap shown in Eq. (1). For all the cases of interest the cross term vanishes, so we omit $[(\partial^2\epsilon/\partial\theta\partial\phi)]_0$ in what follows. Multiple sets of pseudo-Goldstone modes can be handled in a similar fashion, reducing to multiple independent copies of either the type I or type II structures described above (absent fine-tuning).

We have computed the pseudo-Goldstone gap for a wide variety of models [25] using both nonlinear spin-wave theory [Eqs. (5) and (6)], and using the curvature formula [Eq. (1)], which involves only linear spin-wave theory. The results are presented in Table I, where one can see that the two methods agree *exactly* for all models considered. This includes two- and three-dimensional models, isotropic and anisotropic models, models with and without magnon decay, as well as realistic models for two experimental order-by-quantum disorder candidates. Some details for each model, as well as examples of how to define ϕ, θ, and compute the curvatures are provided in the Supplemental Material [25].

Proof of formula.—The equivalence between these two approaches can be understood as follows. The essential ingredient is to notice that the Holstein-Primakoff

TABLE I. Calculations showing the equality of the pseudo-Goldstone gap, Δ, computed from nonlinear spin-wave theory [Eqs. (5) and (6)] and then independently from the curvatures of the classical and quantum zero-point energies [Eq. (1)]. For each model, the lattice, the exchange regime, the type of pseudo-Goldstone mode, and several choice of parameters are listed. When available, additional theoretical or experimental estimates of the pseudo-Goldstone gap are shown.

Model or material	Parameters	Type	Δ	$[(\partial^2\epsilon/\partial\theta^2)]_0$	$[(\partial^2\epsilon/\partial\phi^2)]_0$	$S^{-1}\sqrt{[(\partial^2\epsilon/\partial\theta^2)]_0[(\partial^2\epsilon/\partial\phi^2)]_0}$	$S=\frac{1}{2}$/Exp.
Heisenberg-compass (Square, Ferromagnet)	$\lvert K\rvert/\lvert J\rvert \ll 1$	I	$0.52S^{\frac{1}{2}}\lvert K\rvert^{\frac{3}{2}}/\lvert J\rvert^{\frac{1}{2}}$	$2\lvert K\rvert S^2$	$0.137K^2S/\lvert J\rvert$	$0.52S^{\frac{1}{2}}\lvert K\rvert^{\frac{3}{2}}/\lvert J\rvert^{\frac{1}{2}}$	
	$K/\lvert J\rvert=-0.5$	I	$0.17\lvert J\rvert S^{\frac{1}{2}}$	$\lvert J\rvert S^2$	$0.0286\lvert J\rvert S$	$0.17\lvert J\rvert S^{\frac{1}{2}}$	
Heisenberg-compass [16] (Cubic, Ferromagnet)	$\lvert K\rvert/\lvert J\rvert \ll 1$	II	$0.093K^2/\lvert J\rvert$	$0.093K^2S/\lvert J\rvert$	$0.093K^2S/\lvert J\rvert$	$0.093K^2/\lvert J\rvert$	
	$K/\lvert J\rvert=+0.5$	II	$0.030\lvert J\rvert$	$0.030\lvert J\rvert S$	$0.030\lvert J\rvert S$	$0.030\lvert J\rvert$	
	$K/\lvert J\rvert=-0.5$	II	$0.024\lvert J\rvert$	$0.024\lvert J\rvert S$	$0.024\lvert J\rvert S$	$0.024\lvert J\rvert$	
Heisenberg-Kitaev [60] (Honeycomb, Ferromagnet)	$\lvert K\rvert \ll \lvert J\rvert$	II	$0.0897K^2/\lvert J\rvert$	$0.0897K^2S/\lvert J\rvert$	$0.0897K^2S/\lvert J\rvert$	$0.0897K^2/\lvert J\rvert$	
	$K/\lvert J\rvert=-2.0$	II	$0.208\lvert J\rvert$	$0.208\lvert J\rvert S$	$0.208\lvert J\rvert S$	$0.208\lvert J\rvert$	
	$K/\lvert J\rvert=-0.65$	II	$0.03\lvert J\rvert$	$0.0300\lvert J\rvert S$	$0.0300\lvert J\rvert S$	$0.0300\lvert J\rvert$	$\sim 0.05\lvert J\rvert$ [80]
Heisenberg-Kitaev [60] (Honeycomb, Néel)	$\lvert K\rvert \ll J$	I + I	$0.83\lvert K\rvert S^{\frac{1}{2}}$	$2(3J+K)S^2$	$0.115K^2S/J$	$0.83\lvert K\rvert S^{\frac{1}{2}}$	
	$K/J=+2.0$	I + I	$1.66JS^{\frac{1}{2}}$	$10JS^2$	$0.274JS$	$1.66JS^{\frac{1}{2}}$	
	$K/J=-0.5$	I + I	$0.434JS^{\frac{1}{2}}$	$5JS^2$	$0.038JS$	$0.434JS^{\frac{1}{2}}$	
Heisenberg-Γ [62] (Honeycomb, Ferromagnet)	$\Gamma \ll \lvert J\rvert$	I	$0.29\Gamma^2/\lvert J\rvert\, S^{\frac{1}{2}}$	$3\Gamma S^2$	$0.028\Gamma^3S/\lvert J\rvert^2$	$0.29\Gamma^2/\lvert J\rvert S^{\frac{1}{2}}$	
	$\Gamma/\lvert J\rvert=+0.5$	I	$0.081\lvert J\rvert S^{\frac{1}{2}}$	$1.5\lvert J\rvert S^2$	$0.00437\lvert J\rvert S$	$0.081\lvert J\rvert S^{\frac{1}{2}}$	
	$\Gamma/\lvert J\rvert=+1.0$	I	$0.355\lvert J\rvert S^{\frac{1}{2}}$	$3\lvert J\rvert S^2$	$0.042\lvert J\rvert S$	$0.355\lvert J\rvert S^{\frac{1}{2}}$	
J_1-J_2 [7,11,13] (Square, stripe)	$J_1/J_2 \ll 1$	I + I	$1.44J_1S^{\frac{1}{2}}$	$4(2J_2-J_1)S^2$	$0.2604J_1^2S/J_2$	$1.44J_1S^{\frac{1}{2}}$	
	$J_1/J_2=0.5$	I + I	$0.63J_2S^{\frac{1}{2}}$	$6J_2S^2$	$0.0668J_2S$	$0.63J_2S^{\frac{1}{2}}$	$0.61J_2S^{\frac{1}{2}}$ [81]
	$J_1/J_2=1$	I + I	$1.08J_2S^{\frac{1}{2}}$	$4J_2S^2$	$0.294J_2S$	$1.08J_2S^{\frac{1}{2}}$	$0.96J_2S^{\frac{1}{2}}$ [81]
J_1-J_2 [13,14] (Triangular, stripe)	$J_2/J_1=0.25$	II + II	$0.53J_1$	$0.53J_1S$	$0.53J_1S$	$0.53J_1$	
	$J_2/J_1=0.5$	II + II	$0.45J_1$	$0.45J_1S$	$0.45J_1S$	$0.45J_1$	
	$J_2/J_1=0.75$	II + II	$0.58J_1$	$0.58J_1S$	$0.58J_1S$	$0.58J_1$	
$Er_2Ti_2O_7$ [61,67,68]	Savary *et al.* [61]	I	31.1 μeV	157.5 μeV	1.536 μeV	31.1 μeV	43 − 53 μeV [82,83]
$Ca_3Fe_2Ge_3O_{12}$ [8,65]	Brueckel *et al.* [65]	I + I	262 μeV	4 meV	107.5 μeV	262 μeV	136 μeV [65]

expansion should not depend on the choice of initial classical ground state about which one expands, so long as it is sufficiently close to the true ground state of the model. In other words, the expansion must "self-correct," with the expectation values of the magnons giving the appropriate true ground state spin directions, order by order in $1/S$. For a model without any accidental degeneracies this can be shown at $O(S)$ [25]; the required cancellation relates the magnon energy at zero wave vector to the curvature of the classical energy density, as in the formula of Smit and Beljers [79].

To understand the implication of this self-correction at higher order, we must proceed more indirectly. Define the rotated Hamiltonian $\mathcal{H}(\phi,\theta)=U(\phi,\theta)^\dagger H U(\phi,\theta)$, where $U(\phi,\theta)$ produces the soft configurations of Eq. (7) from the state defined by $\hat{\boldsymbol{z}}_\alpha$. The self-correction condition is then the (trivial) fact that the ground state energy of $\mathcal{H}(\phi,\theta)$ is independent of ϕ and θ. If one considers the implications of this statement on the *derivatives* of the ground state energy of $\mathcal{H}(\phi,\theta)$, at second order one finds that this implies the sum rule [25,84]

$$\boldsymbol{U}_\mu^\dagger\boldsymbol{\sigma}_3\left(\int d\omega\omega\boldsymbol{A}(\boldsymbol{0},\omega)\right)\boldsymbol{\sigma}_3\boldsymbol{U}_\nu=\frac{1}{SN}\left\langle\left(\frac{\partial^2\mathcal{H}}{\partial\lambda_\mu\partial\lambda_\nu}\right)_0\right\rangle, \quad (10)$$

where $\mu,\nu=\Theta,\Phi$, $\lambda_\Phi=\phi$ and $\lambda_\Theta=\theta$ and we define $\boldsymbol{U}_\Phi\equiv(\boldsymbol{V}_0-\boldsymbol{W}_0)/(i\sqrt{2})$, $\boldsymbol{U}_\Theta=(\boldsymbol{V}_0+\boldsymbol{W}_0)/\sqrt{2}$ which span the zero-mode subspace [25]. The magnon spectral function is defined as $\boldsymbol{A}(\boldsymbol{k},\omega)\equiv(2i)^{-1}[\boldsymbol{G}^R(\boldsymbol{k},\omega)-\boldsymbol{G}^A(\boldsymbol{k},\omega)]$, where the $\boldsymbol{G}^A(\boldsymbol{k},\omega)\equiv\boldsymbol{G}^R(\boldsymbol{k},\omega)^\dagger$ is the advanced magnon Green's function.

Using this sum rule [Eq. (10)] one can show that, at $O(S^0)$, the left-hand side is directly related to the pseudo-Goldstone gap, while the right-hand side is related to the curvatures of the classical energy density and quantum zero-point energy density at $O(S^2)$ and $O(S)$, respectively [25]. This argument does not directly extend to higher

orders in $1/S$ or to computing the energies of finite energy modes to $O(S^0)$. We note that in broad strokes this argument bears some resemblance to the Witten-Veneziano formula [85,86] for the mass of the η' meson. In addition, the sum rule [Eq. (10)] is related to Dashen's formula [87] for the mass of pseudo-Goldstone bosons, such as the pion, when chiral symmetry is broken [25].

Discussion.—We now discuss some applications to two leading experimental candidates for order-by-quantum disorder. The first of these is the compound $Ca_3Fe_2Ge_3O_{12}$, which is a three-dimensional $S = 5/2$ version of one of the canonical order-by-disorder models, the $J_1 - J_2$ model [7,13]. This system has a pair of type I pseudo-Goldstone modes, as well as two true Goldstone modes. Because of the low symmetry of the lattice, there are several independent (isotropic) couplings, which have been estimated by comparison of the predictions of linear spin-wave theory with the inelastic neutron scattering spectrum at zero field [65]. One finds that the gap predicted by nonlinear spin-wave theory, $\sim$262 μeV, is of the right order of magnitude, but larger than the 136 μeV [65] observed experimentally [88]. This demonstrates sharply the utility of this method, with the straightforward curvature calculation lending credence to our more involved nonlinear spin-wave result. We also note that the large size of the predicted gap supports the picture that $Ca_3Fe_2Ge_3O_{12}$ is truly an example of order-by-quantum disorder, and perhaps energetic corrections, such as biquadratic interactions [61], are small. This quantitative disagreement could be due to several factors, such as the need to include additional anisotropic or biquadratic exchanges in the model or the need to include interaction or thermal effects in fitting the exchange parameters, which were done at zero magnetic field and moderate temperature [25].

Finally we turn to $Er_2Ti_2O_7$, one of the more ideal material platforms for finding order-by-quantum disorder [61,67]. This is a three-dimensional $S = 1/2$ XY antiferromagnet with a single type I pseudo-Goldstone mode. Using the exchange parameters of Ref. [61], we find that the gap, computed directly in nonlinear spin-wave theory as well as via the curvature formula is $\sim$31 μeV [89]. This theoretical value is still somewhat lower than the 43 μeV [72,82] and 53 μeV [83] that have been reported experimentally in $Er_2Ti_2O_7$. This disagreement could be the result of several factors, such as spin-wave theory being nonquantitative [90] at $S = 1/2$, uncertainties in the exchange parameters [61], or the presence of energetic corrections [70,71] in addition to the order-by-quantum-disorder contribution.

There are many other experimental systems where order-by-quantum disorder may be lurking, and where the results presented here would be useful. In the same vein as $Er_2Ti_2O_7$, order-by-quantum disorder may play a role in the pyrochlores $Yb_2Ti_2O_7$ and its cousin $Yb_2Ge_2O_7$ [91,92], and perhaps even in the ytterbium based spinels [93–96]. Order by disorder has also has played a key role in the understanding of models [60,97,98] of Kitaev materials [23] such as Na_2IrO_3, α-$RuCl_3$ and (α, β, γ)-Li_2IrO_3, and also in related strongly spin-orbit coupled compounds [19,99,100].

On the more theoretical front, one could ask whether the methods discussed could also resolve larger degeneracies, e.g., subextensive line or surface degeneracies [18,101–103]. More drastically, one could consider a case like the kagome antiferromagnet, where the classical ground state is macroscopically degenerate [104–106]. If one expands about states that are expected to be selected by $1/S$ corrections, one finds that the linear spectrum has a large number of zero modes [107]. It would be interesting to compare the semiclassical approach outlined here to the approaches followed in Ref. [108].

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[89] Our result differs from the estimate presented in Ref. [61] by a factor of $\sqrt{2}$. The simplicity of the curvature estimate presented here, relative to that carried out in Ref. [61], as well as the agreement with our explicit nonlinear spin-wave calculation, leads us to trust our value of 31 μeV over the value 22 μeV reported in Ref. [61].
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