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MAGNETIC-FIELD INDUCED SPIN-PEIERLS INSTABILITY
IN STRONGLY FRUSTRATED QUANTUM SPIN LATTICES

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Зумовлена магнітним полем спін-пайєрлсова нестійкість у сильно фрустрованих квантових спінових ґратках

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Анотація. Для класу фрустрованих антиферромагнітних спінових ґраток (зокрема, ґраток квадратне кагомі і кагомі) ми обговорюємо їх стійкість щодо дисторсій, використовуючи недавно знайдені (для цих ґраток) точні власні стани. Ці власні стани є незалежними локалізованими магнонами у ферромагнітному оточенні; вони стають основними станами у сильних магнітних полях. Для відповідних ґраткових дисторсій, припасованих до структури локалізованих магнонів, обчислене точно зменшення магнітної енергії є пропорційне до зміщень атомів, що свідчить про спін-пайєрлсову нестійкість. Оскільки ці локалізовані стани є істотними лише для сильних магнітних полів, ця нестійкість може з'являтися із зміною магнітного поля. Ми обговорюємо також гістерезис спін-пайєрлсового переходу.

Magnetic-field induced spin-Peierls instability in strongly frustrated quantum spin lattices

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Abstract. For a class of frustrated antiferromagnetic spin lattices (in particular, the square-kagomé and kagomé lattices) we discuss the impact of recently discovered exact eigenstates on the stability of the lattice against distortions. These eigenstates consist of independent localized magnons embedded in a ferromagnetic environment and become ground states in high magnetic fields. For appropriate lattice distortions fitting to the structure of the localized magnons the lowering of magnetic energy can be calculated exactly and is proportional to the displacement of atoms leading to a spin-Peierls lattice instability. Since these localized states are present only for high magnetic fields, this instability might be driven by magnetic field. The hysteresis of the spin-Peierls transition is also discussed.

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Antiferromagnetically interacting spin- $\frac{1}{2}$ systems on geometrically frustrated lattices have attracted much attention during last years. Such systems have rich phase diagrams exhibiting a number of unusual quantum phases [1,2]. A striking example is the kagomé lattice antiferromagnet having a liquid-like ground state with a gap for magnetic excitations and a huge number of singlet states below the first triplet state (see e.g. Ref. [3] and references therein). Another intriguing example is the structural phase transition in spin systems driven by magnetoelastic coupling (spin-Peierls instability) observed, e.g., in CuGeO_3 [4]. Frustrating interactions may also provide a route to generating fractional phases in two dimensions which manifest itself in the dynamic correlations probed by inelastic neutron scattering experiments [5].

In the presence of an external magnetic field frustrated quantum spin systems exhibit a number of unusual properties. In particular, plateaus and jumps can be observed in the zero-temperature magnetization curve for such models [1,6]. The theoretical investigation of exotic magnetization curves has been additionally stimulated by the experimental observation of plateaus e.g. in CsCuCl_3 [7] or $\text{SrCu}_2(\text{BO}_3)_2$ [8].

Though the treatment of quantum spin systems often becomes more complicated if frustration is present, in some exceptional cases frustration is crucial to find simple ground states of product form [9,10]. Recently, for a wide class of frustrated spin lattices exact eigenstates consisting of independent localized magnons in a ferromagnetic environment have been found [11,12]. They may become ground states if a strong magnetic field is applied and lead to a macroscopic jump in the zero-temperature magnetization curve just below saturation.

In the present paper we examine the stability of antiferromagnetic spin lattices hosting independent localized magnons with respect to lattice distortions through a magnetoelastic mechanism. We are able to present rigorous analytical results completed by large-scale exact diagonalization data for lattices up to $N = 54$ sites. We discuss the field-tuned changes of the ground-state properties owing to a coupling between spin and lattice degrees of freedom.

The effect of a magnetoelastic coupling in frustrated antiferromagnets is currently widely discussed. First of all the investigation of the Peierls phenomena in frustrated 1D spin systems is in the focus (see, for instance, Ref. [13] and references therein). But also in 2D and 3D quantum spin systems lattice instabilities breaking the translational symmetry are reported. The frustration-driven structural distortions in the spin- $\frac{1}{2}$ square-lattice J_1 - J_2 Heisenberg antiferromagnet studied in Ref. [14] might be relevant for $\text{Li}_2\text{VOSiO}_4$ and VOMoO_4 [14,15]. For 3D frustrat-

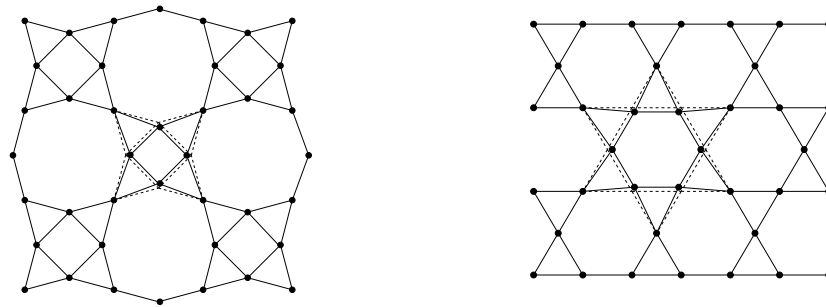


Figure 1. Square-kagomé lattice with one distorted square (left) and kagomé lattice with one distorted hexagon (right) which can host localized magnons. The parts of the lattices before distortions are shown by dashed lines. All bonds in the lattices before distortions have the same length.

ed antiferromagnets containing corner-sharing tetrahedra one discusses several examples for lattice distortions. Inelastic magnetic neutron scattering on ZnCr_2O_4 revealed that a lattice distortion can lower the energy driving the spin system into an ordered phase [16]. NMR investigation of the three-dimensional pyrochlore antiferromagnet $\text{Y}_2\text{Mo}_2\text{O}_7$ gives evidence for discrete lattice distortions which reduce the energy [17]. A lifting of a macroscopic ground-state degeneracy of frustrated magnets through a coupling between spin and lattice degrees of freedom in spin-1 pyrochlore antiferromagnets was studied in Refs. [18,19], which may have relevance to some antiferromagnetic compounds with pyrochlore structure [19].

In all those studies the lattice instability is discussed at zero field. As pointed out already in the late seventies [20] a magnetic field may act against the spin-Peierls transition and might favor a uniform or incommensurate phase. In contrast to those findings, in the present paper we discuss magnetic systems for which the magnetic field is essential for the occurrence of the lattice instability.

To be specific, we consider two geometrically frustrated lattices, namely, the square-kagomé lattice (Fig. 1, left) and the kagomé lattice (Fig. 1, right). The ground state and low-temperature thermodynamics for the Heisenberg antiferromagnet on both lattices are subjects of intensive discussions (see e.g. Refs. [1-3,21,22]). The Hamiltonian of N

quantum ($s = \frac{1}{2}$) spins reads

$$H = \sum_{(nm)} J_{nm} \left(\frac{1}{2} (s_n^+ s_m^- + s_n^- s_m^+) + \Delta s_n^z s_m^z \right) - h S^z. \quad (1)$$

Here the sum runs over the bonds (edges) which connect the sites (vertices) occupied by spins for the lattice under consideration, $J_{nm} > 0$ are the antiferromagnetic exchange constants between the sites n and m , $\Delta \geq 0$ is the anisotropy parameter (in most cases throughout this paper $\Delta = 1$), h is the external magnetic field and $S^z = \sum_n s_n^z$ is the z -component of the total spin. We assume that all bonds in the lattice without distortion have the same length and hence all exchange constants have the same value J .

From Ref. [11] we know that independent localized one-magnon states embedded in a ferromagnetic background are exact eigenstates of the Hamiltonian (1) for the considered models. More specifically, by direct computation one can check that

$$\frac{1}{2} (|\downarrow\downarrow\uparrow\uparrow\rangle - |\uparrow\downarrow\uparrow\uparrow\rangle + |\uparrow\uparrow\downarrow\uparrow\rangle - |\uparrow\uparrow\uparrow\downarrow\rangle) |\dots \uparrow \dots\rangle, \quad (2)$$

$$\frac{1}{\sqrt{6}} (|\downarrow\downarrow\uparrow\uparrow\uparrow\uparrow\rangle - |\uparrow\downarrow\uparrow\uparrow\uparrow\uparrow\rangle + |\uparrow\uparrow\downarrow\uparrow\uparrow\uparrow\rangle - |\uparrow\uparrow\uparrow\downarrow\uparrow\uparrow\rangle + |\uparrow\uparrow\uparrow\uparrow\downarrow\uparrow\rangle - |\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\rangle) |\dots \uparrow \dots\rangle \quad (3)$$

are the one-magnon eigenstates of the Hamiltonian (1) on the square-kagomé and kagomé lattices, where $|\dots \uparrow \dots\rangle$ stands for the embedding fully polarized ferromagnetic environment. The corresponding energies ($h = 0$) of the one-magnon states (2) and (3) are

$$-J + J + (2N - 12) \frac{1}{4} J, \quad (4)$$

$$-\frac{1}{2} J + 2J + (2N - 18) \frac{1}{4} J. \quad (5)$$

The magnons (2) or (3) are trapped (localized) on a square or on a hexagon, respectively. We separate explicitly in (4), (5) the contributions to the energy from those bonds which form a magnon trapping cell (first terms), from the bonds connecting this cell with the environment (second terms) and from the ferromagnetic environment (third terms). One can proceed to fill the lattices with $n > 1$ localized magnons; the state with maximum filling (“magnon crystal”) has $n = n_{\max}$ independent localized magnons with $n_{\max} = \frac{1}{6}N$ and $\frac{1}{9}N$ for the square-kagomé and kagomé lattices, respectively [11,12]. Since S^z commutes with the Hamiltonian

(1), the energy in the presence of an external field $h \neq 0$, $E(S^z, h)$, can be obtained from the energy without field, $E(S^z)$, through the relation $E(S^z, h) = E(S^z) - h S^z$. Since each magnon carries one down spin, a localized magnon state with n independent magnons has $S^z = \frac{1}{2}N - n$ and consequently Eqs. (4), (5) give $E(S^z = \frac{1}{2}N - 1)$ for the corresponding systems.

Under quite general assumptions it was proved [23,24] that these localized magnon states have lowest energies in the corresponding sector of total S^z . As a result, these states become ground states in a strong magnetic field. More specifically, the ground-state energy in the presence of a field is given by $E_0(h) = E_{\min}(S^z) - h S^z$ and the ground-state magnetization S^z is determined from the equation $h = E_{\min}(S^z) - E_{\min}(S^z - 1)$. Since for $S^z = \frac{1}{2}N, \dots, \frac{1}{2}N - n_{\max}$ (i.e., $0 \leq n \leq n_{\max}$) the localized magnon states are the lowest states, one has

$$E_{\min}(S^z) = \frac{1}{2} N J - 3n J = -N J + 3J S^z \quad (6)$$

for both models. Due to the linear relation between E_{\min} and S^z one has a complete degeneracy of all localized magnon states at $h = h_1$, i.e. the energy is $-N J$ at $h = h_1$ for all $\frac{1}{2}N - n_{\max} \leq S^z \leq \frac{1}{2}N$, where $h_1 = 3J$ is the saturation field (identical for both models). Consequently, the zero-temperature magnetization S^z jumps between the saturation value $\frac{1}{2}N$ and the value $\frac{1}{3}N$ ($\frac{7}{18}N$) for the square-kagomé (kagomé) lattice. The effects of the localized magnon states become irrelevant if the spins become classical ($s \rightarrow \infty$).

We want to check the lattice stability of the considered systems with respect to a spin-Peierls mechanism. For this purpose we assume a small lattice deformation which preserves the symmetry of the cell which hosts the localized magnon (in this case the independent localized magnon states remain the exact eigenstates) and analyze the change in the total energy (which consists of the magnetic and elastic parts) to reveal whether the deformation is favorable or not. To find a favorable deformation one needs optimal gain in magnetic energy. For that we use the circumstance that due to the localized nature of the magnons we have an inhomogeneous distribution of nearest-neighbor (NN) spin-spin correlations $\langle \mathbf{s}_i \mathbf{s}_j \rangle$ [12]. In case that one magnon is distributed uniformly over the lattice the deviation of the NN correlation from the ferromagnetic value, $\langle \mathbf{s}_i \mathbf{s}_j \rangle - \frac{1}{4}$, is of the order $\frac{1}{N}$. On the other hand for a localized magnon (3) [(2)] we have along the hexagon [square] hosting the localized magnon actually a negative NN correlation $\langle \mathbf{s}_i \mathbf{s}_j \rangle = -\frac{1}{12}$ [$-\frac{1}{4}$] and all other correlations are positive. Hence a deformation with optimal gain in magnetic energy shall lead to an increase of antiferromagnetic bonds

on the square (hexagon) and to a decrease of the bonds on the attaching triangles. The corresponding deformations are shown in Fig. 1. For the square-kagomé lattice (Fig. 1, left) the deformations lead to the following changes in the exchange interactions: $J \rightarrow (1 + \sqrt{2}\delta) J$ (along the edges of the square) and $J \rightarrow (1 - \frac{1}{4}\sqrt{2}(\sqrt{3} - 1)\delta) J$ (along the two edges of the triangles attached to the square), where the quantity δ is proportional to the displacement of the atoms and the change in the exchange integrals due lattice distortions is taken into account in first order in δ . For the kagomé lattice (Fig. 1, right) one has $J \rightarrow (1 + \delta) J$ (along the edges of the hexagon) and $J \rightarrow (1 - \frac{1}{2}\delta) J$ (along the two edges of the triangles attached to the hexagon). The magnetic energies (4) and (5) are lowered by distortions and become $\frac{1}{2}NJ - 3J - \frac{1}{4}\sqrt{2}(3 + \sqrt{3})\delta J$ and $\frac{1}{2}NJ - 3J - \frac{3}{2}\delta J$, respectively. This is in competition with the increase of the elastic energy, which is given in harmonic approximation by $2(6 - \sqrt{3})\alpha\delta^2$ (square-kagomé) and by $9\alpha\delta^2$ (kagomé). The parameter α is proportional to the elastic constant of the lattice. The change of total energies due to distortions read

$$-\frac{1}{4}\sqrt{2}\left(3 + \sqrt{3}\right)\delta J + 2\left(6 - \sqrt{3}\right)\alpha\delta^2, \quad (7)$$

$$-\frac{3}{2}\delta J + 9\alpha\delta^2, \quad (8)$$

where for n independent localized magnons with n related distortions these results have to be multiplied by n . Minimal total energy is obtained for $\delta = \delta^* = \frac{\sqrt{2}(3+\sqrt{3})J}{16(6-\sqrt{3})\alpha}$ for the square-kagomé and $\delta = \delta^* = \frac{1}{12}\frac{J}{\alpha}$ for the kagomé lattice.

We have considered only a special class of lattice deformations (under which the independent localized one-magnon states survive). Although we are not able to prove rigorously that these lattice deformations are the most favorable, we have presented above nonrigorous arguments that these deformations take advantage of the localized magnons in an optimal way. However, we have rigorously shown that *there exist lattice deformations which yield a gain in the total energy for large values of S^z leading to a spin-Peierls instability of the lattice for an appropriate (large) magnetic field h_1 .*

To discuss the scenario of spin-Peierls instability more specifically we consider a magnetic field above the saturation field h_1 . For the corresponding fully polarized ferromagnetic state a lattice distortion is not favorable. Decreasing h till h_1 the homogeneous ferromagnetic state transforms into the “distorted magnon crystal”; this transformation is accompanied by the aforementioned magnetization jump. On the ba-

sis of general arguments [25,26] we expect that the “magnon crystal” state has gapped excitations and the system exhibits a magnetization plateau between h_1 and $h_2 < h_1$ at $S^z = \frac{1}{2}N - n_{\max}$. To support this statement we calculate the plateau width $\Delta h = h_1 - h_2$ for finite systems of $N = 27, 36, 45, 54$ (kagomé) and $N = 24, 30, 48, 54$ (square-kagomé) for the undistorted lattice, where h_2 is obtained by $h_2 = E_{\min}(S^z = \frac{1}{2}N - n_{\max}) - E_{\min}(S^z = \frac{1}{2}N - n_{\max} - 1)$. Using a $\frac{1}{N}$ finite-size extrapolation we find indeed evidence for a finite $\Delta h = 0.07J$ ($\Delta h = 0.33J$) for the kagomé (square-kagomé) lattice in the thermodynamic limit. This plateau width might be enlarged by distortions (see below).

Now the question arises whether the lattice distortion under consideration is stable below this plateau, i.e., for $S^z < \frac{1}{2}N - n_{\max}$. We are not able to give a rigorous answer but can discuss the question again for finite systems of size $N = 24, 30, 48, 54$ (square-kagomé) and $N = 18, 27, 36, 45, 54$ (kagomé) with n_{\max} distorted squares/hexagons. We calculate the magnetic energy for zero and small distortion parameter δ for different values of S^z . Adopting for the magnetic energy the ansatz

$$E_{\min}(S^z, \delta) = E_{\min}(S^z, 0) + A\delta^p \quad (9)$$

and taking δ of the order of 10^{-4} we can estimate the exponent p from the numerical results. Evidently, the lattice may become unstable if $p < 2$ (and, of course, $A < 0$) whereas $p \geq 2$ indicates lattice stability.

Of course, the numerical results reproduce the analytical findings reported above for $S^z = \frac{1}{2}N - n_{\max}$. More interesting is the sector of S^z just below, i.e. $S^z = \frac{1}{2}N - n_{\max} - 1$. Remarkably both lattices behave differently as h becomes smaller than h_2 . For the finite square-kagomé lattices considered we find $p = 1.001, 1.000, 1.000, 1.000$ for $N = 24, 30, 48, 54$, respectively, if $S^z = \frac{1}{2}N - n_{\max} - 1$. Moreover, p remains equal to 1 for smaller S^z . Therefore, we conclude that the distorted square-kagomé lattice remains stable for $S^z < \frac{1}{2}N - n_{\max}$. On the other hand, for finite kagomé lattices we obtain $p = 2.000, 1.002, 2.000, 1.998, 2.055$ for $N = 18, 27, 36, 45, 54$, respectively, if $S^z = \frac{1}{2}N - n_{\max} - 1$. The “out-rid-er” for $N = 27$ may be attributed to finite-size effects, indeed we have $p = 1.994$ for the next lower $S^z = \frac{1}{2}N - n_{\max} - 2$. Moreover, p remains about 2 for smaller S^z . We interpret small deviations from 2 also as finite-size effects and conclude that the spin-Peierls instability in the kagomé lattice (within the adopted ansatz for the lattice deformation) is favorable only for $\frac{1}{2}N - n_{\max} \leq S^z < \frac{1}{2}N$ and the distortion disappears for $h < h_2$.

The origin for the different behavior of the square-kagomé and the kagomé lattice below h_2 can be attributed to the circumstance, that the square-kagomé lattice has non-equivalent NN bonds but the kagomé lattice has not. This non-equivalency of NN bonds leads to a special affinity of the magnetic system to the considered lattice distortions.

Let us now briefly discuss the influence of the lattice distortion on the saturation field h_1 . Since the fully polarized ferromagnetic state is not distorted, h_1 is shifted to higher values according to $\frac{h_1(\delta^*)}{J} = 3 + \frac{3(2+\sqrt{3})}{32(6-\sqrt{3})} \frac{J}{\alpha}$ for the square-kagomé and $\frac{h_1(\delta^*)}{J} = 3 + \frac{1}{16} \frac{J}{\alpha}$ for the kagomé lattice, i.e. the “distorted magnon crystal” remains stable until $h_1(\delta^*) > h_1$. On the other hand, starting at large magnetic fields $h > h_1$ the fully polarized ferromagnetic state remains (meta)stable until $h_1 = 3J$. Consequently, we have a hysteresis phenomenon in the vicinity of saturation field.

We mention that our considerations basically remain unchanged for the anisotropic Hamiltonian (1) with $\Delta \neq 1$. The presence of anisotropy leads only to quantitative changes in our results. For the sectors of S^z below the localized magnon states we have checked that numerically for $N = 18$ (kagomé) and $N = 24$ (square-kagomé). For the sectors of S^z with localized magnon states it becomes obvious from the change in magnetic energy of a localized-magnon state due to distortions given by $-(\sqrt{2} + \frac{1}{4}\sqrt{2}(\sqrt{3}-1)\Delta)\delta J$ (square-kagomé) and $-(1 + \frac{1}{2}\Delta)\delta J$ (kagomé), where these expressions for $\Delta = 1$ transform to the first terms in Eqs. (7) and (8), respectively.

The spin-Peierls instability in high magnetic fields may appear in other lattices hosting independent localized magnons provided it is possible to construct a lattice distortion preserving the symmetry the localized-magnon cell. Appropriate candidates are e.g. the kagomé-like chain of Ref. [27] or the dimer-plaquette chain (see Ref. [28] and references therein). However, we did not find such a distortion for the sawtooth chain [29,30] or for the kagomé-like chain of Ref. [31].

From the experimental point of view the discussed effect should most spectacularly manifest itself as a hysteresis in the magnetization and the deformation of kagomé-lattice antiferromagnets or the kagomé-like magnetic molecules in the vicinity of the saturation field. Unfortunately, to our best knowledge at the moment such materials are not available (see, however, recent studies [32,33] on a spin- $\frac{1}{2}$ kagomé-like compound).

To summarize, we have reported a spin-Peierls instability in strong magnetic fields for several frustrated Heisenberg antiferromagnets hosting independent localized magnons. This spin-Peierls instability may

or may not survive for smaller fields in dependence on details of lattice structure. In particular, for the Heisenberg antiferromagnet on the kagomé lattice we have found evidence that the spin-Peierls instability breaking spontaneously the translational symmetry of the kagomé lattice appears only in a certain region of the magnetic field. The field dependence of the magnetization and the deformation in the vicinity of saturation displays a hysteresis.

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У СИЛЬНО ФРУСТРОВАНИХ КВАНТОВИХ СПІНОВИХ ГРАТКАХ

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