Macroscopic Magnetization Jumps due to Independent Magnons in Frustrated Quantum Spin Lattices

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For a class of frustrated spin lattices including the Kagomé lattice we construct exact eigenstates consisting of several independent, localized one-magnon states and argue that they are ground states for high magnetic fields. If the maximal number of local magnons scales with the number of spins in the system, which is the case for the Kagomé lattice, the effect persists in the thermodynamic limit and gives rise to a macroscopic jump in the zero-temperature magnetization curve just below the saturation field. The effect decreases with increasing spin quantum number and vanishes in the classical limit. Thus it is a true macroscopic quantum effect.

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In frustrated quantum spin lattices the competition of quantum and frustration effects promises rich physics. A reliable description of such systems often constitutes a challenge for theory. A famous example is the Kagomé lattice antiferromagnet. In spite of extensive studies during the last decade its ground state properties are not fully understood yet. Classically it has infinite continuous degeneracies. In the quantum case ($s = 1/2$), the system is likely to be a spin liquid with a gap for magnetic excitations and a huge number of singlet states below the first triplet state (see [1–3] and references therein).

In this Letter we focus on the zero-temperature magnetic behavior of highly frustrated lattices, in particular, for high magnetic fields. One aspect is given by the observation of nontrivial magnetic plateaus in frustrated two-dimensional (2D) quantum antiferromagnets such as $SrCu_2(BO_3)$ [4,5], which has stimulated theoretical interest (see, e.g., [6]). Also the Kagomé lattice has a plateau at one-third ($m = 1/3$) of the saturation magnetization [7,8]. Since this plateau can be found also in the Ising model and in the classical Heisenberg model with additional thermal fluctuations [9] it can be considered to be of classical origin. However, the structure of the ground state in the classical model is highly nontrivial at $m = 1/3$ [10] and has not been clarified yet for the quantum model.

Another aspect is given by unusual jumps seen in magnetization curves. Such jumps can arise for different reasons. One possibility is a first-order transition between different ground states such as the spin flop transition in classical magnets or in strongly anisotropic quantum chains [11]. Here we discuss another possibility, namely, a macroscopically large degeneracy in the exact ground states of the full quantum system for a certain value of the applied field. We argue that this is a general phenomenon in highly frustrated systems. This is remarkable in so far as one can *exactly* write down ground states at a finite density of magnons in a strongly correlated system which is neither integrable nor has any apparent nontrivial conservation laws. Such jumps represent a genuine macroscopic quantum effect which is also of possible experimental relevance since it occurs in many well-known models such as the Kagomé lattice. This jump occurs just below saturation and should be observable in magnetization experiments on the corresponding compounds if the coupling constants are small enough to make the saturating field accessible.

We consider N quantum spins of "length" s described by the Hamiltonian

$$\hat{H} = \sum_{\langle ij \rangle} J_{ij} \left\{ \Delta \hat{S}_i^z \hat{S}_j^z + \frac{1}{2} (\hat{S}_i^+ \hat{S}_j^- + \hat{S}_i^- \hat{S}_j^+) \right\} - h \hat{S}^z, \quad (1)$$

where the sum runs over neighboring sites $\langle ij \rangle$, $\hat{S}^z$ is the z component of the total spin $\hat{S}^z = \sum_i \hat{S}_i^z$, h is the magnetic field, Δ is the XXZ anisotropy, and J_{ij} are the exchange constants.

If the magnetic field h is sufficiently large, the ground state of (1) becomes the magnon vacuum state $|0\rangle = |\uparrow\uparrow\uparrow \cdots \uparrow\uparrow\uparrow\rangle$ where all spins assume their maximal S_i^z quantum number. The lowest excitations for the case of a high magnetic field are one-magnon states $|1\rangle$. They are represented by states where the S^z quantum number is lowered by 1 and can be written as

$$|1\rangle = \frac{1}{c} \sum_i^N a_i \hat{S}_i^- |0\rangle, \quad (2)$$

where c is chosen such that $\langle 1 | 1 \rangle = 1$. For a lattice with n_s spins per unit cell, there are n_s possible magnon bands $w_i(\vec{k})$ with $i = 0, \ldots, (n_s - 1)$. For certain combinations of J_{ij} of the Hamiltonian (1) the lowest magnon dispersion $w_0(\vec{k})$ becomes flat (i.e., independent of k) in some directions or in the whole k space.

If the one-magnon dispersion is independent of one of the components of $\vec{k}$, one can use Fourier transformation along this direction to localize the one-magnon excitation along this direction in space. If the dispersion is completely flat, the magnon can be localized in a finite region of the lattice. This localized excitation can have N/n_s different positions. Now it is clear that one can construct further local excitations. There will be no interaction with the other excitations as long as they are sufficiently well separated in space, and therefore each excitation will have the same energy. In this manner, one obtains n-magnon excitations for $n \leq n_{\max}$ whose energy is exactly n times the one-magnon energy. Because of the absence of attractive interaction, it is plausible that these excitations are also the lowest n-magnon excitations. A proof of this statement for $s = 1/2$, arbitrary $\Delta \geq 0$, and all J_{ij} equal will be given elsewhere [12]; below we report numerical evidence for the occurrence of this effect for several models. The essence of the above argument is that the ground state energies in the $1, 2, \ldots, n_{\max}$ magnon spaces depend linearly on the number of magnons, i.e., on the total magnetic quantum number. Hence the total S^z in the ground state goes directly from $Ns - n_{\max}$ to the saturation value Ns when increasing the magnetic field. In terms of the magnetization curve $m(h) = S^z(h)/(Ns)$, this implies that there is a jump $\delta m = n_{\max}/(Ns)$. If one band is completely flat, the system can support a macroscopic amount of independent magnons $n_{\max} \sim N$ and one obtains a macroscopic jump just below saturation.

To be more precise, denote the region of localization of the magnon state by L. Then the coefficients a_l in (2) are different from zero only for sites $l \in L$. The local one-magnon state is completely decoupled from the rest of the lattice R and the eigenstate $|\Psi\rangle$ can be written as a product $|\Psi\rangle = |\Psi_L\rangle|\Psi_R\rangle$ of a local part L and the rest R. $|\Psi_L\rangle$ is the local magnon state and $|\Psi_R\rangle$ is the vacuum state. The coefficients a_i vanish for $i \in R$ in the one-magnon state (2); i.e., $a_i \neq 0 \; \forall \, i \in L$ and $a_i = 0 \; \forall \, i \in R$.

The necessary and sufficient condition for decoupling of the local state from the rest R is

$$\sum_{l \in L} a_l J_{lk} = 0 \quad \forall \; k \in R \, . \tag{3}$$

The Hamiltonian (1) can be divided into three parts,

$$\hat{H} = \hat{H}_L + \hat{H}_{L-R} + \hat{H}_R \, , \tag{4}$$

with L being the part of the lattice where one magnon is localized and R the rest. The first term $\hat{H}_L$ is the local part of the Hamiltonian with $J_{ij} = J_{l_1 l_2}$ and $l_1, l_2 \in L$, whereof $|\Psi_L\rangle$ is the lowest eigenstate. The second term $\hat{H}_{L-R}$ is the coupling of the local part to the rest of the lattice with $J_{ij} = J_{lk}$ and $l \in L$, $k \in R$. J_{lk} must satisfy condition (3). The rest of the Hamiltonian which is not connected with the local part is $\hat{H}_R$ with $J_{ij} = J_{k_1 k_2}$ and $k_1, k_2 \in R$. $\hat{H}_{L-R}$ together with condition (3) creates the frustration; therefore, it seems that the magnetization jump described here is restricted to highly frustrated lattices. The simplest realizations of such a Hamiltonian are rings connected only by triangles.

In the Kagomé lattice [1,7] as a typical example of flat one-magnon dispersion $w_0(\vec{k}) = h - 2sJ(1 + 2\Delta)$ the magnon can be localized around a hexagon (see Fig. 1). Choosing the coefficients $a_l = (-1)^l$ with l numbering the sites around a hexagon ensures that $|\Psi_L\rangle$ is an exact eigenstate for the hexagon (as illustrated by the bold one in Fig. 1). The triangles around the hexagon fulfill condition (3) and therefore the magnon on the hexagon is decoupled from the rest of the lattice, showing that this state is also an eigenstate of the whole Kagomé lattice. Further magnons can be put on the lattice without disturbing existing excitations. This can be repeated until every third hexagon is excited as shown in Fig. 1. As a consequence we have a macroscopic magnetization jump δm having its maximal value $\delta m = 2/9$ for the extreme quantum case $s = 1/2$.

A proof that these states are the lowest eigenstates in the corresponding sectors is given in [12] for $s = 1/2$, arbitrary $\Delta \geq 0$ and all J_{ij} equal. Here we present numerical evidence for this statement. In Fig. 2 exact diagonalization results are shown for finite systems with periodic boundary conditions for the $s = 1/2$ Heisenberg antiferromagnet on the Kagomé lattice. Only lattices with N being a multiple of 9 (three unit cells) are presented, which fit to the $m = 7/9$ state corresponding to Fig. 1. The jump to saturation can easily be seen in this figure. We have also computed curves on smaller clusters. They agree with those presented in [7,8]. If the boundary conditions do not fit to the $\sqrt{3} \times \sqrt{3}$ state or if the cluster is too small, the jump may show finite-size effects.

We emphasize that the existence of this jump is quite independent of several details of the system. First, one can construct $N/9$ independent magnons for any s, leading to a jump of height $\delta m = 1/(9s)$. Indeed, the results for the $s = 1$ Kagomé lattice presented in [7] show a jump of the expected height for the given cluster sizes. Second,

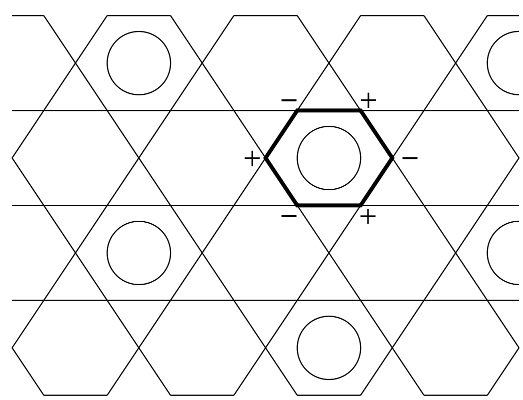

FIG. 1. Kagomé spin array, which hosts at least $N/9$ independent magnons in a $\sqrt{3} \times \sqrt{3}$ structure if it consists of N spins. The circles mark hexagons where the independent magnons can be localized. The structure of one localized magnon is indicated by the signs around the bold hexagon which correspond to coefficient $a_l = \pm 1$ with which a spin flip contributes at each site l. Together with the surrounding triangles the local state satisfies Eq. (3).

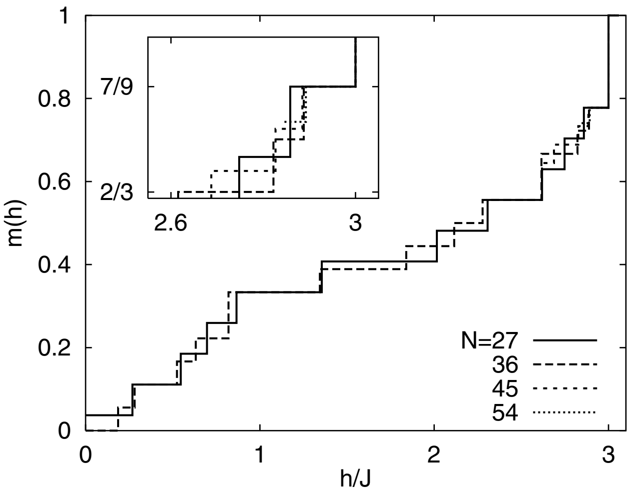

FIG. 2. Magnetization curves of an $s = 1/2$ Heisenberg antiferromagnet on a finite Kagomé lattice for $N = 27, 36, 45, 54$ and $\Delta = 1$. For $N = 45$ and $N = 54$ the curve starts just below the magnetization jump. It can be clearly seen that the jump to full magnetization has *no* finite size dependence.

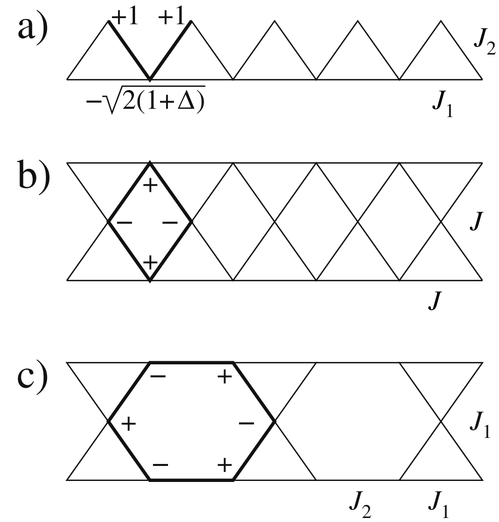

FIG. 3. One-dimensional quantum antiferromagnets with a magnetization jump to the full moment $m = 1$: (a) the sawtooth chain [15,16], (b) the Kagomé-like chain of [17], and (c) the Kagomé-like chain of [18]. The position of a local magnon is marked by thick lines with coefficients a_l attached.

introduction of an XXZ anisotropy $\Delta \neq 1$ does not affect the crucial properties of the one-magnon dispersion, and therefore one expects the magnitude of the degeneracy and the associated jump to be independent of Δ. In the exact diagonalization results for $s = 1/2$ [8] jumps of identical size are indeed observed for $\Delta = 0$, 1, and 2.5. Third, the argument remains also unchanged if one generalizes to different coupling constants in the triangles pointing up and down (see Fig. 1) [3]. The degeneracy is (partially) lifted only if coupling constants are changed such that they become different around one triangle. The jump therefore seems to be very stable not only in the Kagomé lattice but also in the other models to be discussed next where similar arguments can be applied.

Another 2D example for completely flat one-magnon dispersion $w_0(\vec{k}) = h - 2sJ(1 + 3\Delta)$ is the checkerboard lattice [13], a 2D variant of the pyrochlore lattice. In this case, localized magnon excitations live around a square without diagonal interactions, again with coefficients $a_l = (-1)^l$. The magnetization jump is $\delta m = 1/(8s)$. We have verified the predicted degeneracy and associated macroscopic jump numerically for the checkerboard lattice with $s = 1/2$ and $\Delta = 1$.

Completely flat bands can also be found in dimensions different from two. For example, the generalized pyrochlore lattice in three dimensions with two different coupling constants J, J' (see, e.g., [14]) gives rise to the high frustration necessary for a decoupling of local magnon excitations. The lowest two out of the four magnon bands are indeed degenerate and completely flat: $w_0(\vec{k}) = w_1(\vec{k}) = h - s(J + J')(1 + 3\Delta)$. We expect a macroscopic jump of $\delta m \geq 1/(12s)$ for all $J, J' \geq 0$.

Even in one dimension, one can find systems with a flat dispersion: Some examples are shown in Fig. 3 together with the structure of the localized magnon excitations. For the generalized sawtooth chain [15,16] of Fig. 3(a), the lowest magnon branch is completely flat for $J_2 = \sqrt{2(1 + \Delta)}\, J_1$ and all s. Note that this example satisfies (3) with more complicated coefficients a_l than encountered previously. The lowest magnon branch for the one-dimensional Kagomé variant [17] shown in Fig. 3(b) is also completely flat: $w_0(\vec{k}) = h - 2sJ(1 + 2\Delta)$. Figure 3(c) shows another variant of a Kagomé chain [18]. Here, the state indicated by the bold hexagon is an eigenstate for $J_2 = (2\Delta + 1)J_1/(\Delta + 1)$ with $w_0(\vec{k}) = h - 2sJ_1(1 + 2\Delta)$. We have checked numerically that, for $s = 1/2$ and $\Delta = 1$, 0, a jump of size $\delta m = 1/2$, $1/3$, or $1/5$ exists in cases (a), (b), or (c), respectively. As an example, Fig. 4 shows the $m(h)$ curve of the model of Fig. 3(c) with $s = 1/2$, $\Delta = 0$, and $J_2 = J_1$ (to ensure a flat dispersion). The jump of height $\delta m = 1/5$ can be seen clearly (compare also the inset). Furthermore, one can see several plateaus in the magnetization curve. This suggests that the same conditions which give rise to the jump also favor the formation of magnetization plateaus. In particular, our numerical data always show a plateau preceding the jump. However, this is beyond the scope of the present Letter and needs further investigations.

So far, we have discussed cases with completely flat dispersion. However, in more than one dimension it is also possible that the lowest magnon branch has a flat dispersion only in some, but not all directions. This has been noticed previously for the J_1-J_2 model on the square lattice antiferromagnet with $J_2 = J_1/2$ [19]. The generalized checkerboard lattice [13] is another 2D model which has flat directions if the couplings J' along the diagonals are larger than those for the square lattice J. In these cases,

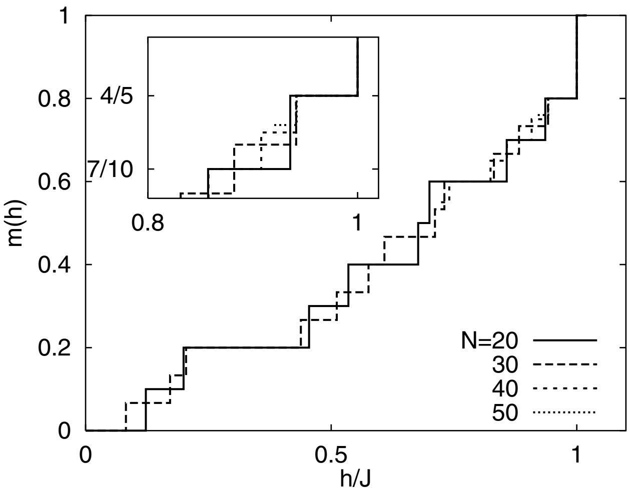

FIG. 4. Magnetization curves of the $s = 1/2$ XY-Kagomé chain ($\Delta = 0$) of Fig. 3(c) with $J_1 = J_2 = J$ for $N = 20$, 30, 40, 50. Inset: region just below the jump to saturation.

magnons remain delocalized along one dimension and thus the localization region L is a one-dimensional object. For the J_1-J_2 model, L extends along one of the axes of the square lattice while for the checkerboard model it extends along one of the diagonals. In both cases, $a_l = (-1)^l$ satisfies the condition (3) and yields localized magnon excitations. However, now the number of possible magnon excitations is bounded by the linear extent of the lattice and therefore their density tends to zero in the thermodynamic limit. Consequently, the jump is observable only in finite systems (this has been checked for both models with $s = 1/2$ and $\Delta = 1$) and vanishes in the thermodynamic limit. Arguments similar to those used in [20] then indicate that all derivatives $d^i h(m)/dm^i$ with $i \geq 1$ vanish at the point of full magnetization $m = 1$. This is in contrast to [20] where $d^2 h(m)/dm^2$ was assumed to be nonzero and thus a square root was inferred in $m(h)$ for the J_1-J_2 model with $J_2 = J_1/2$. Since the transition appears to become continuous for $N \to \infty$, a nonanalytic functional dependence in $h(m)$ like $h(m) \sim \exp[-1/(1 - m)]$ appears more plausible. In any case, also in such a situation the magnetization curve will be exceptionally steep close to saturation, a fact which should also be detectable in high field experiments on materials realizing such models.

In summary, we have shown for certain frustrated spin lattices that for magnetic fields near saturation noninteracting magnons can condensate into a single-particle ground state leading to a macroscopic jump in the magnetization curve. This is a true quantum effect which vanishes if the spins become classical ($s \to \infty$).

Since this effect is generic in highly frustrated magnets, we are confident that a realization of some model discussed in the present Letter can be found with sufficient small s and J to make the saturating field accessible. The dynamics of the lattice may be relevant in such a compound, but one will have to see whether this strengthens or weakens the anomaly predicted at the saturating field. Our analysis of deformations of the coupling constants at least indicates that the low-temperature magnetization curve will show an unusually steep rise even if the geometry deviates from the ideal structure. It is interesting to note that a similar jump also appears in certain molecular magnets [12] where the magnetic ions form an icosidodecahedron (such as $\{Mo_{72}Fe_{30}\}$), a cuboctahedron, or similar frustrated structures.

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Note added in proof.—Recently, one of the present authors generalized the proof that the states constructed in this Letter are ground states to general s [21].

[1] P. Lecheminant, B. Bernu, C. Lhuillier, L. Pierre, and P. Sindzingre, Phys. Rev. B **56**, 2521 (1997).
[2] Ch. Waldtmann, H.-U. Everts, B. Bernu, C. Lhuillier, P. Sindzingre, P. Lecheminant, and L. Pierre, Eur. Phys. J. B **2**, 501 (1998).
[3] F. Mila, Phys. Rev. Lett. **81**, 2356 (1998).
[4] H. Kageyama, K. Yoshimura, R. Stern, N. V. Mushnikov, K. Onizuka, M. Kato, K. Kosuge, C. P. Slichter, T. Goto, and Y. Ueda, Phys. Rev. Lett. **82**, 3168 (1999).
[5] K. Onizuka, H. Kageyama, Y. Narumi, K. Kindo, Y. Ueda, and T. Goto, J. Phys. Soc. Jpn. **69**, 1016 (2000).
[6] T. Momoi and K. Totsuka, Phys. Rev. B **62**, 15 067 (2000).
[7] K. Hida, J. Phys. Soc. Jpn. **70**, 3673 (2001).
[8] D. C. Cabra, M. D. Grynberg, P. C. W. Holdsworth, and P. Pujol, Phys. Rev. B **65**, 094418 (2002).
[9] M. E. Zhitomirsky, A. Honecker, and O. A. Petrenko, Phys. Rev. Lett. **85**, 3269 (2000).
[10] M. E. Zhitomirsky, Phys. Rev. Lett. **88**, 057204 (2002).
[11] C. Gerhardt, K.-H. Mütter, and H. Kröger, Phys. Rev. B **57**, 11 504 (1998).
[12] J. Schnack, H.-J. Schmidt, J. Richter, and J. Schulenburg, Eur. Phys. J. B **24**, 475 (2001).
[13] B. Canals, Phys. Rev. B (to be published), cond-mat/0102233.
[14] B. Canals and C. Lacroix, Phys. Rev. Lett. **80**, 2933 (1998).
[15] T. Nakamura and K. Kubo, Phys. Rev. B **53**, 6393 (1996).
[16] D. Sen, B. S. Shastry, R. E. Walstedt, and R. Cava, Phys. Rev. B **53**, 6401 (1996).
[17] Ch. Waldtmann, H. Kreutzmann, U. Schollwöck, K. Maisinger, and H.-U. Everts, Phys. Rev. B **62**, 9472 (2000).
[18] P. Azaria, C. Hooley, P. Lecheminant, C. Lhuillier, and A. M. Tsvelik, Phys. Rev. Lett. **81**, 1694 (1998).
[19] A. Honecker, Can. J. Phys. **79**, 1557 (2001).
[20] M. S. Yang and K. H. Mütter, Z. Phys. B **104**, 117 (1997).
[21] H.-J. Schmidt, cond-mat/0203270.