Macroscopic magnetization jumps due to independent magnons in frustrated quantum spin lattices

J. Schuleenburg, A. Honecker, J. Schnack, J. Richter and H.-J. Schmidt

1 Institut für Theoretische Physik, Universität Magdeburg, P.O. Box 4120, D-39016 Magdeburg, Germany
2 Institut für Theoretische Physik, TU Braunschweig, Mendelssohnstr. 3, D-38106 Braunschweig, Germany and
3 Universität Osnabrück, Fachbereich Physik, Barbarastr. 7, D-49069 Osnabrück, Germany

(Dated: August 29, 2001; revised February 25, 2002)

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.

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. 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, 2, 3] and references therein). 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 classical model is highly non-trivial for high magnetic fields. One aspect is given by the observation of nontrivial magnetic plateaus in frustrated two dimensional (2D) quantum antiferromagnets like SrCu$_2$(BO$_3$)$_2$ [4, 5], which has stimulated theoretical interest (see e.g. [6]).

In this Letter we will 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 like SrCu$_2$(BO$_3$)$_2$ 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 it can be considered to be of classical origin. However, the structure of the ground state in the classical model is highly non-trivial at m=1/3 and has not been clarified yet for the quantum model.

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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 like 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 non-trivial 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 like 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

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

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

If the magnetic field $h$ is sufficient large, the ground state of $\hat{H}$ becomes the magnon vacuum state $|0\rangle = | \uparrow\uparrow\uparrow \ldots \uparrow\uparrow\uparrow \rangle$ 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 $S_i^z$-quantum number is lowered by one and can be written as

$$
|1\rangle = \frac{1}{c} \sum_{i=1}^{N} a_i \hat{S}_i^- |0\rangle,
$$

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 $\hat{H}$, 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
The Hamiltonian (1) can be divided into three parts $H \equiv H_L + R + H_R$ with $L$ being the part of the lattice where one magnon is localized and $R$ the rest. The first term $H_L$ is the local part of the Hamiltonian with $J_{ij} = J_{1,1_L}$ and $l_1, l_2 \in L$, whereof $|\Psi_L\rangle$ is the lowest eigenstate. The second term $H_{L-R}$ is the coupling of the local part to the rest of the lattice with $J_{ij} = J_{ik}$ and $l \in L, k \in R$. The rest of the Hamiltonian which is not connected with the local part is $H_R$ with $J_{ij} = J_{k_1,k_2}$ and $k_1, k_2 \in R$. $H_{L-R}$ 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.
We have verified the predicted degeneracy and associated jump to be independent of $\Delta$. 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. Some examples are shown in Fig. 3 together with the structure of the localized magnon excitations. For the generalized sawtooth chain $\Delta$ of Fig. 3a), the lowest magnon branch is completely flat for $J_2 = \sqrt{2(1+\Delta)}J_1$ and all $\Delta$. 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 shown in Fig. 3b) is also completely flat $w_0(\vec{k}) = h - 2sJ(1+2\Delta)$. Fig. 3c) shows another variant of a kagomé chain $w_0(\vec{k}) = h - 2sJ(1+2\Delta)$. 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 case a), b) or c), respectively. As an example, Fig. 4 shows the $m(h)$ curve of the model of Fig. 3a) 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 to a jump of height $\delta m = 1/(9s)$. Indeed, the results for the $s = 1$ kagomé lattice presented in Fig. 4 show a jump of the expected height for the given cluster sizes. Second, 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 $\Delta$. In the exact diagonalization results for $s = 1/2$ jumps of identical size are indeed observed for $\Delta = 0$, $1$ and $2.5$. Third, the argumentation remains also unchanged if one generalizes to different coupling constants in the triangles pointing up and down (see Fig. 1). 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, 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., [3]) 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 with the structure of the localized magnon excitations. For the generalized sawtooth chain $\Delta$ of Fig. 3a), the lowest magnon branch is completely flat for $J_2 = \sqrt{2(1+\Delta)}J_1$ and all $\Delta$. 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 shown in Fig. 3b) is also completely flat $w_0(\vec{k}) = h - 2sJ(1+2\Delta)$. Fig. 3c) shows another variant of a kagomé chain $\Delta$. 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 case a), b) or c), respectively. As an example, Fig. 4 shows the $m(h)$ curve of the model of Fig. 3a) 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.

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lattices that for magnetic fields near saturation non-interacting 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 (like \{Mo$_7$Fe$_3$\}), a cuboctahedron or similar frustrated structures.

Acknowledgement: This work was partly supported by the DFG (project Ri615/10-1).

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