Self-consistent spin wave analysis of the magnetization plateau in triangular antiferromagnet

J Takano¹, H Tsunetsugu¹ and M E Zhitomirsky²,³

¹Institute for Solid State Physics, University of Tokyo, Kashiwanoha 5-1-5, Kashiwa, Chiba 277-0882, Japan
²SPSMS, UMR-E9001, CEA-INAC/UJF, 17 rue des Martyrs, 38054 Grenoble Cedex 9, France
³Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer str. 38, D-01187 Dresden, Germany

E-mail: takano@issp.u-tokyo.ac.jp

Abstract. We investigate theoretically the 1/3 magnetization plateau of the triangular-lattice Heisenberg antiferromagnet at zero temperature. Using the self-consistent spin-wave analysis, we show that the 1/3 plateau is stabilized in a finite range of magnetic fields: $0.84 < H < 1.39$ for $S = 1/2$. By analyzing the Bose condensation of magnons, we demonstrate that the magnetization curve has logarithmic singularities at the both ends of the plateau and determine the prefactor of the singularity up to the leading order in $1/S$.

1. Introduction
The Heisenberg antiferromagnet on a triangular lattice has been intensively studied in the past as the basic example of geometric frustration in magnetism. One of the characteristic features of the triangular antiferromagnet is the magnetization process. Existence of the 1/3 magnetization plateau with spins forming the up-up-down structure has been theoretically established by spin wave [1], exact diagonalization [2], and coupled cluster expansion [3] studies. Recently, the 1/3-magnetization plateau has been observed in several magnetic materials, $\text{Cs}_2\text{CuBr}_2$ [4] with $S = 1/2$, $\text{RbFe(MoO}_4)_2$ [5] and $\text{Rb}_4\text{Mn(MoO}_4)_3$ [6] with $S = 5/2$. However, there is a quantitative discrepancy between analytical and numerical results about the plateau transition points. Chubukov et al. evaluated the plateau region in the leading order of $1/S$ as $1 - 0.084/S < H/3SJ < 1 + 0.215/S$ [1]. The exact diagonalization study [3] yields $0.92 < H/3SJ < 1.44$ for $S = 1/2$. While the upper plateau boundary is given rather accurately by the $1/S$ expansion, there is substantial discrepancy in the prediction of the lower transition point. Moreover, nature of the transitions at the endpoints of the plateau and corresponding critical behavior are not well understood by either analytical or numerical studies.

To improve quantitative understanding of the 1/3 plateau, we perform the self-consistent spin wave analysis in the plateau phase. We derive the plateau transition points, excitation spectrum inside the plateau phase and a singularity of magnetization curve at both ends of the plateau. Interaction between bosons has an essential role to stabilize the plateau phase, and we take it into account in the following two steps. First, we perform Hartree-Fock approximation on bosonic Hamiltonian defined in the vacuum of up-up-down state. We calculate the width of magnetization plateau phase with gapped excitations, which is qualitatively the same as the
previous study based on $1/S$ expansion [1]. Second, we analyze the singularity of the plateau transition. By taking successive magnon-magnon scatterings into account, we show a logarithmic singularity of magnetization curve at the endpoints of the plateau. Finally we compare our results with the other studies.

2. Self-consistent calculation in up-up-down vacuum

First we derive a bosonic Hamiltonian by Holstein-Primakoff transformation with the vacuum of up-up-down configuration, and analyze it using Hartree-Fock approximation. We obtain excitation spectrum with a finite energy gap, which is a linear function of magnetic field. We also determine the position of plateau transition by calculating the magnetic field where the gap closes.

We study the antiferromagnetic Heisenberg model on a triangular lattice in magnetic field

$$
\mathcal{H} = J \sum_{\langle ij \rangle} S_i \cdot S_j - 3SJ h \sum_i S_i^z. \quad (J = 1)
$$

(1)

Note that classically the magnetization curve is straight and the up-up-down configuration is stable only at $h = 1$. To bosonize this Hamiltonian, we use the Holstein-Primakoff transformation introducing three types of boson operators

$$
S_i^z = -S + b_i^A, \quad S_i^+ = -b_i^A \sqrt{2S - b_i^A b_i^A}, \quad S_i^- = -\sqrt{2S - b_i^A b_i^A} b_i^A, \quad i \in A \text{ sublattice}
$$

$$
S_i^z = S - b_i^B, \quad S_i^+ = \sqrt{2S - b_i^B b_i^B} b_i^B, \quad S_i^- = b_i^B \sqrt{2S - b_i^B b_i^B}, \quad i \in B \text{ sublattice}
$$

(2)

Second we perform Hartree-Fock decoupling on quartic terms to derive an effective quadratic Hamiltonian. To do this, we introduce various kinds of boson-pair expectation values, which are self-consistently determined:

$$
n_a \equiv \langle b_i^a b_i^a \rangle, \quad m_{aa'} \equiv \langle b_i^a b_j^{a'} \rangle, \quad \Delta_{aa'} \equiv \langle b_i^a b_j^{a'} \rangle,
$$

(3)

where

$$
\nu_k = \frac{1}{3} \left( e^{ik\delta_1} + e^{ik\delta_2} + e^{ik\delta_3} \right),
$$

(4)

with lattice vectors $\delta_1 = (1,0), \delta_2 = (-1/2, \sqrt{3}/2), \delta_3 = (-1/2, -\sqrt{3}/2)$ and $(a, a') = (A,B), (B,C), (C,A)$. Some of these are related to each other and we use these relations here without explicit explanation. It is important to notice that the mean-field Hamiltonian commutes with total $S^z$ operator and therefore with the Zeeman term. The mean-Field Hamiltonian can be thus decoupled as

$$
\mathcal{H}_{HF} = 3S \sum_k \alpha_k^+ \left[ \hat{\mathcal{H}}_0(k) - \delta h \hat{\Lambda} + \frac{1}{S} \hat{\mathcal{H}}_1(k) \right] \alpha_k + \text{const.}, \quad \alpha_k = (b^A_k, b^{B1}_k, b^{C1}_k)^T.
$$

(5)

Here $\hat{\mathcal{H}}_0(k)$ is quadratic term at magnetic field $h = 1$

$$
\hat{\mathcal{H}}_0(k) = \begin{pmatrix}
1 & -\nu_k & -\nu_k \\
-\nu_k & 1 & \nu_k \\
-\nu_k & \nu_k & 1
\end{pmatrix}.
$$

(6)

$-\delta h \hat{\Lambda}$ is the remaining Zeeman energy with $\delta h = h - 1$, $\hat{\Lambda} = \text{diag}(1,-1,-1)$. $\hat{\mathcal{H}}_1$ denotes the mean field term

$$
\hat{\mathcal{H}}_1(k) = \begin{pmatrix}
-2(n_B - \Delta CA) \\
\frac{1}{2}(n_A + n_B - 2\Delta CA)\nu_k \\
\frac{1}{2}(n_A + n_B - 2\Delta CA)\nu_k
\end{pmatrix} + \begin{pmatrix}
\frac{1}{2}(n_A + n_B - 2\Delta CA)\nu_k \\
\frac{1}{2}(n_A + n_B - 2\Delta CA)\nu_k \\
\frac{1}{2}(n_A + n_B - 2\Delta CA)\nu_k
\end{pmatrix}.
$$

(7)
We use the Bogoliubov transformation and diagonalize the quadratic Hamiltonian (5) as

\[ \mathcal{H}_{HF} = 3S \sum_k \sum_{j=1}^3 \omega_j(k) \beta_j^{\dagger} \beta_j. \]  

(8)

It is important that the Bogoliubov transformation is independent of magnetic field \( h \) and three modes consist of one \( S^+ \) mode \((j = 2)\) and two \( S^- \) modes \((j = 1, 3)\), as the original boson operators defined by (2). Therefore, the only effect of applied field in the plateau phase is uniform shift of three magnon branches:

\[ \omega_{1,3}(k) = \omega_{1,3}^{(h=1)}(k) + \delta h, \quad \omega_2(k) = \omega_2^{(h=1)}(k) - \delta h. \]  

(9)

Plateau transition points \( h_{c1} \) and \( h_{c2} \) \((h_{c1} < h_{c2})\) are defined by the magnetic field where the lowest mode touches energy zero

\[ h_{c1} = 1 - \omega_1^{(h=1)}(k = 0), \quad h_{c2} = 1 + \omega_2^{(h=1)}(k = 0). \]  

(10)

Note that the two transition points are not symmetric about \( h = 1 \) due to the difference of energy gaps of the two modes.

Finally we calculate the spectrum and estimate the plateau transition points. We solve the self-consistent equations numerically and derive the magnon spectra \( \{\omega_j(k)\}\). We show the spectra for the \( S = 1/2 \) case in fig.1. They are given by the absolute values of the three roots of the cubic equation

\[ x^3 + 0.778x^2 + (-1.921 + 1.654|\nu_k|^2)x - 1.723 + 4.072|\nu_k|^2 - 1.203(\nu_k^2 + \nu_k^3) = 0, \]  

(11)

where one positive root corresponds to \( S^+ \) mode \((j = 2)\) and two negative roots correspond to \( S^- \) modes \((j = 1, 3)\).

Plateau transition points are calculated as explained above and the results are

\[ (h_{c1}, h_{c2}) = \begin{cases} 
(0.844, 1.368) & S = 1/2 \\
(0.920, 1.195) & S = 1 
\end{cases}. \]  

(12)

The plateau width is thus 0.524 for \( S = 1/2 \). This is quite improved towards the numerical value compared with the \( 1/S \) expansion result, although the values of transition points themselves are not so precise.
3. Analysis of plateau transition

We now analyze singularity of the plateau transitions. These transitions can be viewed as Bose condensation of one of the bosonic excitations derived in the previous section. We take into account successive magnon scatterings, which has dominant contribution to the self energy of the boson near the transition point, and show the presence of logarithmic singularity of magnetization.

When the energy gap closes, the Hartree-Fock solution derived in the previous section becomes unstable and Bose condensation occurs. This physically means the onset of canting of spin configuration. Our goal is to calculate the density of condensed bosons asymptotically correctly. To describe this singularity, we consider a state slightly outside the plateau phase and set $h = h_{c2} \pm \mu$, $j = 1, 2 \mu \ll 1$. Since only the lowest mode $\omega_j$ is relevant to the critical behavior, we can neglect the other two modes and regard the system as consisting of only one type of bosons which have kinetic energy $\omega_j^{(k=h_{c2})}(k) - \mu$. For convenience, we consider an equivalent alternative model: the ground canonical ensemble with chemical potential $\mu$ and gapless dispersion relation $\omega_j(k) - \omega_j(0) \equiv \epsilon(k)$. With this set-up, the density of condensed bosons is determined by Hugenholtz-Pines relation [7]

$$\Sigma_{11}(k = 0, \omega = 0) - \Sigma_{12}(k = 0, \omega = 0) = \mu, \quad (13)$$

where $\Sigma_{11}$ and $\Sigma_{12}$ denote normal and anomalous self energy, respectively. Note that this chemical potential $\mu$ is defined including the factor $1/3S$. As is clear from fig.1, the dispersion relation is quadratic near $k = 0$, and can be written as $\epsilon(k) \sim ak^2$.

Let us consider which terms in the self energy are dominant. Ladder type contribution shown in fig.2(a) diverges as $k \to 0$, $\omega \to 0$, and we have to correctly take their contribution into account to determine the critical behavior. Moreover, the Green’s function in fig.2(a) should be calculated self-consistently to avoid a divergence, and it is necessary to take the terms illustrated in fig.2(b) into account. This approximation is also shown as fig.2(c), using vertex part determined by the Bethe-Salpeter equation fig.2(d). Hereafter we analyze the asymptotic behavior of magnetization as $\mu \to 0$, which is determined by the self-consistent equation for self energy fig.2(c)(d) and Hugenholtz-Pines relation (13) in the large $S$ limit.

We further simplify the Green’s functions in fig.2(d). Since the Green’s function with small $k$ and $\omega$ has dominant contribution in the vertex part, the self energy may be replaced by the limiting value:

$$\Sigma_{11}(k, \omega) \to \Sigma_{11}(0, 0), \quad \Sigma_{12}(k, \omega) \to \Sigma_{12}(0, 0). \quad (14)$$

Moreover, as $\mu \to 0$, the normal vertex part shown in fig.2(c) predominates over the anomalous vertex part fig.2(f), and we neglect the latter. As a result, the normal and anomalous self energies determined by fig.2(c) satisfy $\Sigma_{11}(0, 0) = 2\Sigma_{12}(0, 0)$. Combining this with (13), we obtain

$$\Sigma_{11}(0, 0) = 2\mu, \quad \Sigma_{12}(0, 0) = \mu. \quad (15)$$

These self energies are to be used to evaluate the Green’s function in the vertex part fig.2(d).

Then we evaluate the normal vertex part with zero momentum, $\Gamma$. The Bethe-Salpeter equation shown in fig.2(d) is solved as

$$\Gamma = \frac{\Gamma^{(0)}}{1 - i\Gamma^{(0)} \sum_k \frac{d\omega}{2\pi} G_{11}(k, \omega)G_{11}(-k, -\omega)}, \quad (16)$$

where the zeroth-order vertex is approximated by a constant:

$$\Gamma^{(0)}(k_1, k_2, k_3, k_4) = \Gamma^{(0)}(0, 0, 0, 0). \quad (17)$$
Figure 2. (a) Ladder type contribution to boson self energy. Dotted line denotes condensed boson at $k = 0$. (b) Ladder type contribution with self-consistently determined Green’s function. (c) Alternative expression of (b) using vertex part. (d) Bethe-Salpeter equation which determines the vertex part in (c). (e) Normal component of vertex part. (f) Anomalous component of vertex part.

The zeroth-order vertex $\Gamma^{(0)}$ can be calculated from the original bosonic Hamiltonian represented in terms of Bogoliubov quasiparticles derived in the previous section, and the result is $\Gamma^{(0)} = 3/SN$. Although the integral in (15) has an infrared divergence, we can cancel out this divergence by including the contribution of the anomalous Green’s function $G_{12}$:

$$\int \frac{d\omega}{2\pi} [G_{11}(k, \omega) + G_{12}(k, \omega)] [G_{11}(-k, -\omega) + G_{12}(-k, -\omega)].$$

The resulting vertex part $\Gamma$ contains both the normal and anomalous parts. But we regard $\Gamma$ as the normal part, since the contribution from the latter is negligible as $\mu \to 0$. Explicit calculation yields

$$\Gamma = \frac{3}{SN} \left( 1 - \frac{3\sqrt{3} \ln \mu}{16\pi S a} \right)^{-1}.$$  

Finally we determine the number of condensed bosons $N_0$ and then determine the singularity of magnetization. Self energy is determined by the vertex part as shown in fig.2(c) and we obtain

$$\Sigma_{11}(0) = 2N_0 \Gamma = \frac{6N_0}{SN} \left( 1 - \frac{3\sqrt{3} \ln \mu}{16\pi S a} \right)^{-1}.$$  

Combining this with the first relation in (15), we obtain the number of condensed bosons:

$$N_0 = \frac{N}{3} \left( S \mu - \frac{3\sqrt{3} \mu \ln \mu}{16\pi a} \right).$$

The change of the ground state energy due to condensation is given by

$$\Delta E = 3S \left[ -\mu N_0 + \frac{1}{2} \Gamma N_0^2 \right].$$
The change of magnetization per site is calculated by differentiating this as

$$\Delta M = \pm \frac{1}{3} \left[ -S\mu + \frac{3\sqrt{3}\mu \ln \mu}{16\pi a} \right].$$  \hspace{1cm} (23)

Where the upper(lower) sign corresponds to $h_{c1}(h_{c2})$. Inverse mass of excitation spectrum $a$ can be calculated with the quadratic Hamiltonian (6) and the result is $a = 1/4 + O(1/S)$ for $h = h_{c1}$ and $a = 3/4 + O(1/S)$ for $h = h_{c2}$. Therefore the change of magnetization is

$$\Delta M/S = \begin{cases} 
\frac{1}{3} \Delta h + \frac{\sqrt{3}}{4S\pi} \Delta h \ln \Delta h & \text{at } h = h_{c1}, \\
\frac{1}{3} \Delta h - \frac{\sqrt{3}}{12S\pi} \Delta h \ln \Delta h & \text{at } h = h_{c2},
\end{cases}$$  \hspace{1cm} (24)

where $\Delta h = h_{c1} - h$ or $h - h_{c2}$.

This logarithmic singularity of magnetization curve generally appears near the saturation field of two-dimensional antiferromagnet [8], and has been studied through introducing infrared cutoff [9, 10], or using renormalization group [11]. Our result (24) agrees with those results up to the prefactor, if the boson mass is properly set.

4. Conclusion

In this paper we have performed spin wave analysis on the 1/3 magnetization plateau of the triangular-lattice Heisenberg antiferromagnet. We have proved that collinear configuration guarantees that the energy gap vanishes completely linearly in magnetic field. Our Hartree-Fock calculation predicted the transition points $H/(3JS) = 0.844$ and 1.368. Although there exist some discrepancies between those values and numerical results, the plateau width is substantially improved compared with the $1/S$ expansion. Regarding the singularity at the plateau transitions, we have demonstrated the presence of logarithmic term by taking account of magnon multiple scatterings. This agrees with common behavior of bose condensation in two dimensions, but this has not been predicted for this problem yet, within the knowledge of the authors. We have succeeded in reproducing the correct logarithmic term up to prefactor by means of a method which is much simpler than other approaches. This may be also useful for further improvement by considering higher order terms.

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