Rotons and spin transport in square lattice antiferromagnets in magnetic fields

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Abstract. We investigate the excitation spectrum and spin conductivity in square lattice antiferromagnets by nonlinear spin-wave theory. We have found that the excitation spectrum have a rotonlike minimum at about 3/4 of the saturation field. The “roton” appears and softens rapidly to zero as a function of the magnetic field as a result of the especially strong three-magnon interactions. For the spin conductivity, it becomes clear that a two-magnon sideband appears for sufficiently strong three-magnon interactions.

1. Introduction
In zero magnetic field, square lattice Heisenberg antiferromagnets (SLHAFs) have the Néel structure and three-magnon interactions [1–4] are prohibited. Accordingly, an excitation spectrum at zero field is in good agreements with linear spin-wave (LSW) results with small renormalization [5].

In finite magnetic fields, SLHAFs have noncollinear canted states and three-magnon interactions are known to come into play [1–4]. Moreover, three-magnon interactions can be tuned from zero to quite large values by the external magnetic field. Contrary to the collinear Néel structure, sizable deviations from LSW calculations are pointed out for SLHAFs in high fields both numerically [6, 7] and experimentally [8]. Moreover, nonlinear spin-wave spectra are shown to become negative at some wavevectors in high fields [3].

Motivated by Ref. [3], we investigate the nonlinear spin-wave spectra within the second-order perturbation calculations based on the formalism of Zhitomirsky-Nikuni-Chernyshev [1, 2, 4]. We focus on the field region just below where we get the negative magnon spectra, near 3/4 of the saturation field, and we find a rotonlike minimum on the acoustic mode which softens rapidly to zero with small increase of fields [9]. We believe that the complete softening of the roton has crucial importance, and a new ground state should appear in higher magnetic fields.

We also study the spin conductivity in SLHAFs in fields by the linear response theory since it is expected that the appearance of a “roton” should change thermal and transport properties rather drastically. We compare the results of the LSW calculations [10] and the one with 1/S corrections. A two-magnon sideband appears with 1/S corrections in high fields as a result of the strong three-magnon interactions. We have checked that f-sum rule [10] is satisfied even after the emergence of the sideband.
2. Model
We summarize only the main points of the spin-wave formalism of Zhitomirsky-Nikuni-Chernyshev [1, 2, 4]. Hamiltonian $\hat{H}$ of SLHAF in the magnetic field $H$ is given by

$$\hat{H} = J \sum_{<i,j>} (S_i^{x_0} S_j^{x_0} + S_i^{y_0} S_j^{y_0} + S_i^{z_0} S_j^{z_0}) - H \sum_i S_i^z,$$

(1)

where $S_i^{\mu_0} (\mu_0 = x_0, y_0, z_0)$ denotes the spin operator of a laboratory frame, and $J$ is the exchange constant. We move from the laboratory frame to a rotating frame

$$S_i^x = \frac{S_i}{\Phi} \sin \theta + S_i^z e^{iQ \cdot r_i} \cos \theta, \quad S_i^y = S_i^z, \quad S_i^z = -S_i^x e^{iQ \cdot r_i} \cos \theta + S_i^z \sin \theta,$$

(2)

where $S_i^{\mu} (\mu = x, y, z)$ stands for the spin operator of the rotating frame, $\theta$ is the canting angle, and $Q = (\pi, \pi)$ is the ordering wave vector.

We perform the Holstein-Primakoff (HP) transformation to $\hat{H}$ with bosons $a_i$:

$$S_i^+ = \sqrt{2S - a_i^+ a_i} \quad a_i, \quad S_i^- = a_i^+ \sqrt{2S - a_i^+ a_i}, \quad S_i^z = S - a_i^+ a_i.$$

(3)

We get the HP series of the Hamiltonian: $\hat{H} = \sum_n \hat{H}_n$, where $\hat{H}_n (n = 0, 1, 2, \cdots)$ represents the term $n$-th order in HP bosons.

We determine the classical canting angle $\theta$ by the condition:

$$\frac{\partial \hat{H}_0}{\partial \theta} = 0.$$  

(4)

We get $\theta$ as

$$\theta = \arcsin [h], \quad h = H/H_s, \quad H_s = 8JS,$$

(5)

where $H_s$ denotes the saturation field and $\hat{H}_1$ vanishes by determining $\theta$ correctly.

We then perform the Fourier transformation and then the Bogoliubov transformation with bosons $b_k$:

$$a_k = u_k b_k + v_k b_k^\dagger, \quad (u_k^2 - v_k^2 = 1).$$

(6)

We get the linear spin-wave energy from $\hat{H}_2$

$$\omega_k = 4JS \sqrt{(1 + \gamma_k)(1 - \cos 2\theta \gamma_k)}, \quad \gamma_k = \frac{\cos k_x + \cos k_y}{2}.$$

(7)

At zero field, there are two massless Goldstone-bosons at the $\Gamma$ point and the $M = (\pi, \pi)$ point. For finite fields, however, the Goldstone-boson at the $\Gamma$ point becomes massive while that of the $M$ point remains massless. The curvature near the $M$ point increases monotonically as the field increases, and it changes the sign for $h \geq h^* = 2/\sqrt{7}$ as mentioned in Refs. [2–4].

3. Nonlinear Spin-Wave Theory
We start from Zhitomirsky-Nikuni-Chernyshev nonlinear interaction Hamiltonian [1, 2, 4]

$$\hat{H}_3 = \sum_{k,q} \Phi_1(k, q, p_1)(b_k^\dagger b_{p_1} b_q + H.c.) + \sum_{k,q} \Phi_2(k, q, p_2)(b_k^\dagger b_{p_2} b_q^\dagger + H.c.),$$

(8)
to discuss their effects on spin-wave spectra by the second-order perturbation theory. For brevity, we put $p_1 = Q + k - q$, $p_2 = Q - k - q$. The coupling constants $\Phi_n(k, q, p_n) [n = 1, 2] \propto \sin 2\theta$ are given in Refs. [2, 3]. The three-magnon interaction term $\mathcal{H}_3$ comes into play only in the noncollinear structure [1–4], and the strength is tunable by the magnetic field in SLHAFs.

The on-shell self-energies due to the three-magnon interactions are

$$
\Sigma_1(k, \omega_k) = \frac{1}{2} \sum_q \frac{\Phi_1(k, q, p_1)^2}{\omega_k - \omega_{p_1} - \omega_q + i\delta},
\Sigma_2(k, \omega_k) = -\frac{1}{2} \sum_q \frac{\Phi_2(k, q, p_2)^2}{\omega_k + \omega_{p_2} + \omega_q - i\delta},
$$

We see from Eq. (9) that the three-magnon interactions $\Sigma_1(k, \omega_k)$ become strong where the energy differences of the initial- and intermediate-states are small. For $h \geq h^* = 2/\sqrt{7}$, the spontaneous magnon decay becomes possible near the $M$ point, and the three-magnon interactions are expected to become strong near the decay threshold [2–4].

In addition, we must consider Hartree-Fock decoupling of $\mathcal{H}_4$ and quantum corrections of the canting angle when we calculate the nonlinear spin-wave spectrum $\tilde{e}_k$ [2–4].

We also study the imaginary part of the excitation spectra [3]:

$$
\Gamma_k = \frac{\pi}{2} \sum_q \Phi_1(k, q, p_1)^2 \delta(\omega_k - \omega_{p_1} - \omega_q),
$$

and calculate the spectral weight [9]:

$$
A(k, \omega) = \frac{1}{\pi} \frac{\Gamma_k}{(\omega - \epsilon_k)^2 + \Gamma_k^2},
$$

4. Linear response theory

The main points of the linear response theory [11–14] for the spin conductivity is summarized below. The spin current density $J_s(\omega)$ induced by a field gradient $\nabla_x h(\omega)$ is written as:

$$
J_s = \sigma_s(\omega) \nabla_x h(\omega),
$$

where $\sigma_s(\omega)$ is the spin conductivity. We get a Lehman representation of the regular parts of the long wavelength spin conductivity at zero temperature [13]:

$$
\sigma_{s,\text{reg}}(\omega) = \frac{\pi}{N} \sum_m \frac{|\langle m | j_s(q) | 0 \rangle|^2}{\omega} \delta(\omega - E_m + E_0) \delta(\omega + E_m - E_0),
$$

where $j_s(q)$ denotes the spin current density operator in the Fourier representation.

Here, the spin conductivity $\sigma_s(q, \omega)$ can be related to dynamical structure factor [3]:

$$
S_{\omega_0, \omega_0}(q, \omega) = \int_{-\infty}^{\infty} \frac{dt}{2\pi} \langle S_{q, \omega_0}^{\omega_0} S_{-q, \omega}^{\omega_0} \rangle e^{i\omega t}
$$

by following relation [15, 16]:

$$
\sigma_s(q, \omega) = \frac{\omega}{4\sin^2(q_L)} S_{\omega_0, \omega_0}(q, \omega),
$$

as long as the magnetization is conserved. We now see the importance to study the spin conductivity since it is can be related to experimentally observable quantity $S_{\omega_0, \omega_0}(q, \omega)$. 


Detailed derivations of the spin current density operators \(j_{s,i,i+\hat{x}}\) of noncollinear magnets, using the conservation of the magnetization, are given in Ref. [10]. In the present paper, we only show \(j_{s,i,i+\hat{x}}\) of SLHAFs using spin operators of the laboratory frame:

\[
j_{s,i,i+\hat{x}} = J \left( S_i^x a^{\dagger}_i S_{i+\hat{x}}^{\dagger,0} - S_i^{\dagger,0} S_{i+\hat{x}}^x \right).
\]

(16)

Here, we calculate spin conductivity by nonlinear spin-wave theory. We move from the laboratory frame to the rotating frame, and perform HP expansion. We get

\[
\bar{j}_{s,i,i+\hat{x}} = \sum_n \bar{j}_{s,i,i+\hat{x}}^{(3-n)/2} \quad (n = 0, 1, 2, \cdots),
\]

(17)

where \(\bar{j}_{s,i,i+\hat{x}}^{(3-n)/2}\) is the part of the spin current operator of \((n-3)/2\)-th order in \(1/S\).

We calculate the spin conductivity by the second-order perturbation theory, and we need spin operators:

\[
\bar{j}_{s,i,i+\hat{x}}^{3/2} = -iJS \sqrt{\frac{S}{2}} e^{\frac{3iS}{4}a^{\dagger}_i} \cos \theta \left[ (a_i - a^{\dagger}_i) + (a_{i+\hat{x}} - a^{\dagger}_{i+\hat{x}}) \right],
\]

(18)

\[
\bar{j}_{s,i,i+\hat{x}}^{1/2} = -iJS \left( a^{\dagger}_i a_{i+\hat{x}} - a_i a^{\dagger}_{i+\hat{x}} \right) \sin \theta,
\]

(19)

\[
\bar{j}_{s,i,i+\hat{x}}^{1/2} = iJ \sqrt{\frac{S}{2}} e^{\frac{3iS}{4}a^{\dagger}_i} \cos \theta \left[ (a_i - a^{\dagger}_i) n_{i+\hat{x}} + (a_{i+\hat{x}} - a^{\dagger}_{i+\hat{x}}) n_i \right]
\]
\[
+ n_i a_i - a_i^{\dagger} n_i + n_{i+\hat{x}} a_{i+\hat{x}} - a_{i+\hat{x}}^{\dagger} n_{i+\hat{x}}\right].
\]

(20)

We see that the nonlinear contribution \(\bar{j}_{s,i,i+\hat{x}}^{1/2}\) vanishes in the collinear structure at zero field, and comes into play only in finite fields. We show later the strong nonlinearity lead to a two-magnon sideband.

5. Appearance of a roton
Nonlinear spin-wave spectra \(\tilde{\epsilon}_k\) \((S = 1/2)\) in various fields are shown in Figure 1(a) [9]. The spectra change drastically for small changes of fields at around \(h \sim 0.75\). Enlarged spin-wave spectra \(\tilde{\epsilon}_k\) near the point \(M\) is shown in Fig. 1(b) [9]. We see that a rotonlike minimum appears on the acoustic mode. It becomes clear that this roton is truly a minimum in the Brillouin zone [9].

We use small value \(\delta/4JS = 2.5 \times 10^{-6}\) to calculate Eq. (9), and used Mathematica for calculation. We obtain qualitatively the same results for any reasonable values of \(\delta\) but roton gap become larger for larger \(\delta\). We also add that we chose \(\delta\) as small as possible within the range where a numerical integration is convergent.

Interestingly, this roton’s shape changes drastically and roton gap drops rapidly to zero with a slight increase of the field [9]. In other words, this roton is tunable by the external field, which should cause drastic changes of thermal and transport properties.

The spectral weight \(A(k, \omega)\) at \(h = 0.7565\) is shown in Fig. 2 (a) and at \(h = 0.7568\) in Fig. 2 (b) [9]. We colored red for \(A(k, \omega)\) higher than 20. We now see that the roton feature and its softening survive even after considering the magnon decay. We use Lorentzian to approximate the delta function of the imaginary part of the spectra [Eq. (10)], and used \(\zeta/4JS = 6 \times 10^{-5}\) for small values of the Lorentzian. Only more spread imaginary parts \(\Gamma_k\) are obtained for larger \(\zeta\) and qualitatively the same results are expected for any reliable \(\zeta\). Here, we put \(\zeta\) in the same way as how we chose \(\delta\).

Since the roton appears in extremely narrow regions of magnetic fields and wavevector rather resonantly, we can expect that the complete roton softening remains even after considerations of
higher order corrections including four-magnon interactions. We also believe that the complete softening of the roton [9] and the negative spectra in higher fields [3] means that the simple canted state is no longer the ground state in high fields. Therefore, it is our future work to investigate the new ground state in higher fields.

In noncollinear frustrated antiferromagnets, the appearance of a rotonlike minimum has already been pointed out both experimentally [17–19] and theoretically [20, 21]. We emphasize that there seems to have been no reports on rotons, to our knowledge in other systems, which changes drastically and softens rapidly with small increases of the magnetic field [9]. We expect the roton in SLHAFs can be detected by neutron, thermal experiments, details are given in Ref. [9], and Raman scattering. It can also be detected in the spin conductivity by a two-roton sideband, which appears about twice the energy of the roton gap. We discuss the result of the spin conductivity within the second-order perturbation theory in the next section.

6. Strong Nonlinear Effects on Spin Conductivity
We now discuss the nonlinear effects on the spin conductivity for SLHAFs ($S = 1/2$) in fields with the second-order perturbation theory. The details of the LSW results of the spin conductivity are already given in Ref. [10]. In the present paper, we compare the spin conductivities calculated with or without $1/S$ corrections. The spin conductivity at zero field is shown in Figure 3(a), and only small quantitative changes are observed. The threshold of the spin conductivity, which originates from the van-Hove singularity [10, 12], shifts by taking $1/S$ corrections into account.
Figure 3. (a) Spin conductivities ($S = 1/2$) calculated within LSW (dashed black line) and the one calculated with $1/S$ corrections (red line) at zero field. No qualitative changes are observed by taking $1/S$ corrections into account. (b) The spin conductivity at $h = 0.125$ is shown. We also show the conductivity with $1/S$ corrections mediated by one-magnon intermediate states (dashed and dotted blue line) and two-magnon intermediate states (dotted blue line). (c) The spin conductivity at $h = 0.7$. A two-magnon sideband appears by strong three-magnon interactions. (d) The spin conductivity at $h = 0.75$. We see that the two-magnon sideband grows as the field increases.

This is due to the hardening of the spectrum by Hartree-Fock decoupling [7, 22]. We also see the suppression of the spin conductivity with $1/S$ corrections, which originates from the reduction of the ordered moment by quantum fluctuations [6, 10]. The spin conductivity at $h = 0.125$ is shown in Fig. 3(b). A two-magnon structure, which vanishes in the collinear structure, appears as a result of the three-magnon interactions.

The spin conductivity at $h = 0.7$ and $h = 0.75$ are shown in Fig. 3(c) and Fig. 3(d). A two-magnon sideband appears as a result of the strong three-magnon interactions, and grows as the field increases. We have checked that the $\ell$-sum rule [10] is approximately satisfied even after the emergence of the two-magnon sidebands. We also see a narrowing of the bandwidth, which is attributed to the negative contribution of $\Sigma_1(k, \omega)$. Unfortunately, we do not see the effect of the roton feature in this approximations. We need to take higher order $1/S$ corrections into account to examine the effects of the roton [23].

The rotonlike minimum also appears in frustrated noncollinear magnets [20, 21]. Possible effects of the roton on the two-magnon Raman spectrum are examined previously in frustrated antiferromagnets [18, 19, 23]. These works suggest the emergence of a two-roton Raman spectrum, which appears about twice the roton gap. We see that the frequency shifts of two-roton sideband in spin conductivity might also be observed by taking higher order $1/S$ corrections.

In contrast to the frustrated magnets, rotons in SLHAFs are shown to be very sensitive to the magnetic field [9]. This implies that the two-roton Raman spectrum should show the corresponding sharp dependence on the field: rather sudden appearance of the two-roton feature in the two-magnon Raman spectrum near $h = 3/4$, followed by the rapid softening of the two-roton spectrum with a small change of the field. This softening should provide a direct and strong evidence to the existence of rotons in SLHAFs.
7. Conclusion
We have shown that the effects of the three-magnon interactions on the nonlinear spin-wave spectrum and the spin conductivity by the second-order perturbation theory. We have found that the roton appears just below the field region to get negative magnon spectra [3] as a result of the strong three-magnon interactions. The roton shape changes and the roton gap drops rapidly to zero with a small increase of the field at around $h \sim 0.75$ [9]. We believe that we get positive excitation spectra again by finding the new ground states in higher fields, and it is our future work to reveal the new ground state.

We have also studied the spin conductivity in SLHAFs. Significant changes are observed in high fields by taking $1/S$ corrections into account. The two-magnon sideband appears and it grows as the field increases.

We expect to observe the two-roton structure in the Raman spectrum and spin conductivity, in addition to the neutron and thermal measurements [9], showing the rapid shift of the spectrum. We believe that this might become a conclusive evidence of the existence of the roton. Last, it is our future work to examine the two-magnon Raman spectrum in SLHAFs in fields.

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