Spin conductivity in two-dimensional non-collinear antiferromagnets

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We propose a method to derive the spin current operator for non-collinear Heisenberg antiferromagnets. We show that the spin conductivity calculated by the spectral representation with the spin current thus derived satisfies the f-sum rule. We also study spin conductivity at $T = 0$ within spin wave theory. We show how spin conductivity depends on external magnetic field with changing magnon spectrum. We also find that spin Drude weight vanishes for any external magnetic field at $T = 0$.

KEYWORDS: spin current, antiferromagnet, f-sum rule, Drude weight, linear response theory

1. Introduction

Spin currents have attracted considerable interest with development of spintronics in recent years. Spin conductivity is well studied theoretically in one dimensional antiferromagnets by many methods including exact diagonalization.$^{1,2}$ It is also studied in two dimensional antiferromagnets.$^{3,4}$ These theories on spin currents, however, are rather restricted to collinear antiferromagnets. As far as the authors are aware, there seems to be no clear definition of the spin current operator in the case of non-collinear antiferromagnets.

One of our main purposes is to introduce a definition of the spin current operator for non-collinear quantum antiferromagnets; typical examples include Heisenberg antiferromagnets in square and triangular lattices under static homogeneous magnetic fields.$^{5-7}$ Spins cant on each sublattice with magnetic fields, but still the spin current operator can be defined by using equation of continuity as we show explicitly in this paper.

This letter is composed as follows. First, we show a way to define the spin current operator for non-collinear quantum antiferromagnets. We consider $S = 1/2$ Heisenberg spins on square and triangular lattices to illustrate our method which, we believe, is applicable to much wider classes of non-collinear antiferromagnets. We transform from a laboratory frame to a rotating frame for convenience.$^{5,6}$ We then introduce a Holstein-Primakoff boson common to all sublattices, and diagonalize the harmonic part of the Holstein-Primakoff expansion by

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Bogoliubov transformation. We employ the linear response theory in spectral representation to calculate spin conductivity, which is found to satisfy f-sum rule. We find that the Drude weight vanishes. We also show how the magnon excitation spectrum affects the frequency dependence of spin conductivity when external magnetic field is varied.

2. Spin waves in Rotating Frame

We now propose a method to define spin wave operators in non-collinear antiferromagnets. First, we introduce a useful spin wave formalism of non-collinear Heisenberg antiferromagnets on square and triangular lattices. We suppose each spin on different sublattices to be on the \( x_0 - z_0 \) plane of the laboratory frame \((x_0, y_0, z_0)\), and transform it to the rotating frame \((x, y, z)\) for the square lattice

\[
S^{30}_j = e^{iQ \cdot r_j} S^z_j \cos \theta + S^x_j \sin \theta
\]

\[
S^{30}_j = S^y_j
\]

\[
S^{20}_j = S^z_j \sin \theta - e^{iQ \cdot r_j} S^x_j \cos \theta,
\]

and for the triangular lattice

\[
S^{30}_j = S^z_j \sin \theta_j + S^x_j \cos \theta_j
\]

\[
S^{30}_j = S^y_j
\]

\[
S^{20}_j = S^z_j \cos \theta_j - S^x_j \sin \theta_j.
\]

We stress that spins on each sublattice are now expressed by a simple set of rotated spin operators \( S^\mu_i (\mu = x, y, z) \) common to all sublattices.

Next, we consider uniform magnetic fields \( h \leq 8JS \) on square lattice, and \( h \leq 3JS \) on triangular lattice. Magnetization saturates at \( h = 8JS \) in square lattice, and spins select up-up-down phase at \( h = 3JS \) in triangular lattice. The model spin Hamiltonian for both lattices are written in the same form in the laboratory frame

\[
\hat{H} = J \sum_{\langle i,j \rangle} (S^{x0}_i S^{x0}_j + S^{30}_i S^{30}_j + S^{20}_i S^{20}_j) - h \sum_i S^{z0}_i.
\]

Canting angles are determined to make the ground state energy \( E_0 \) minimum. For square lattice, it is given by

\[
\theta = \sin^{-1} \left( \frac{h}{8JS} \right).
\]
Canting angles for triangular lattice are given by\(^7\)
\[
\begin{align*}
\theta_A &= -\pi \\
\theta_B &= -\theta_C \\
\theta_B &= \cos^{-1}\left[\frac{1}{2} \left(\frac{h}{3JS} + 1\right)\right] (h \leq 3JS),
\end{align*}
\]
for sublattice A, B, C. We perform Holstein-Primakoff transformations to spin operators on the rotating frame\(^5,6\)
\[
\begin{align*}
S_j^+ &= \sqrt{2S - a_j^\dagger a_j} \\
S_j^- &= a_j^\dagger \sqrt{2S - a_j^\dagger a_j} \\
S_j^z &= S - a_j^\dagger a_j.
\end{align*}
\]
We perform Fourier transformation, and then Bogoliubov transformation
\[
a_k^\dagger = u_k b_k^\dagger + v_k b_{-k}
\]
to diagonalize the harmonic part of the bosonic Hamiltonian.\(^5,6\) In this way, we get spin-wave spectrum for square lattice\(^5\)
\[
\omega_k = 4JS \sqrt{(1 + \gamma_k)(1 - \gamma_k \cos 2\theta)}
\]
\[
\gamma_k = \frac{1}{2}(\cos k_x + \cos k_y),
\]
and for triangular lattice\(^6,7\)
\[
\omega_k = 3JS \sqrt{(1 + 2\gamma_k) \left(1 + \gamma_k \left(\frac{h}{3JS}\right)^2 - 1\right)}
\]
\[
\gamma_k = \frac{1}{3} \left(\cos k_x + 2 \cos \frac{k_x}{2} \cos \frac{\sqrt{3}k_y}{2}\right).
\]
We see that the energy gap \(\Delta = h\) opens at \(\Gamma\) point in both lattices when an external magnetic field \(h\) is applied. We derive a spin current operator for Heisenberg antiferromagnets in the next section.

3. Spin currents

3.1 Derivation of a spin current operator

Now, we derive spin current operators \(J_s\) for both lattices by spin wave operators given in the preceding section. Spin conductivity \(\sigma_s(\omega)\) is defined by linear response relation
\[
J_s = \sigma_s \nabla_s h, \tag{10}
\]
when $\nabla_x h$ is the gradient of magnetic fields along the $x$–direction and $\mathcal{J}_s$ is the induced spin current.\textsuperscript{1–4} We assume that spin current flows along the field gradient. We can use equation of continuity to derive a spin current operator, because magnetization is a conserved quantity in Heisenberg antiferromagnets. Equation of continuity in the long wavelength limit for spin current density

$$J_s = \sum_i j_{s,i,i+x}$$

is written as\textsuperscript{1–3}

$$\frac{1}{\Omega N} \frac{\partial}{\partial t} S_z^0_i = -j_{s,i,i+x} - j_{s,i,i-x} a_0,$$ \hspace{1cm} (12)

where $\Omega$ denotes area of unit cell, $N$ denotes number of lattice sites and $a_0$ denotes lattice constant. We get a spin current operator by Heisenberg equation of motion

$$\frac{\partial}{\partial t} S_z^0_i = \{\hat{H}, S_z^0_i\},$$

$$J_s = \frac{a_0}{\Omega} \sum_i \{\hat{H}_{i,i+x}, S_z^0_i\},$$

$$= \frac{a_0}{\Omega} \sum_i J \left( S_{i,i}^{z0} S_{i,i+x}^{z0} - S_{i,i}^{z0} S_{i,i+x}^{Tentative} \right).$$ \hspace{1cm} (13)

Then, we transform from the laboratory frame to the rotating frame in each lattice and perform Holstein-Primakoff expansion. We get the spin current operator

$$J_s = \sum_n J_{s,n/2} \quad n = 3, 2, 1 \cdots,$$ \hspace{1cm} (14)

where $J_{s,n/2}$ is a term proportional to $S^{n/2}$. For the square lattice, the leading term $J_{s,3/2}$ is given by

$$J_{s,3/2} = \frac{-ia_0JS}{\Omega} \sqrt{\frac{S}{2}} \sum_i e^{iQ \cdot R_i} \cos \theta \left( a_i - a_i^+ \right),$$

$$= \frac{-ia_0JS}{\Omega} \sqrt{\frac{S}{2}} \sum_i e^{iQ \cdot R_i} \cos \theta \left( a_{i+x} - a_{i+x}^+ \right),$$ \hspace{1cm} (15)

and for the triangular lattice, it is given by

$$J_{s,3/2} = \frac{ia_0JS}{\Omega} \sqrt{\frac{S}{2}} \sum_i \sin \theta_{i,i+x} \left( a_i - a_i^+ \right),$$

$$= \frac{ia_0JS}{\Omega} \sqrt{\frac{S}{2}} \sum_i \sin \theta_i \left( a_{i,i+x} - a_{i,i+x}^+ \right).$$ \hspace{1cm} (16)

We see that spin current operators $J_{s,3/2}$ are first order in bosonic operators.
3.2 Spin conductivity

Next, we show how we calculate spin conductivity \( \sigma_s(\omega) \), which is written by the Drude weight \( D_s \) and the regular part \( \sigma_{s,\text{reg}}(\omega) \)

\[
\sigma_s(\omega) = D_s \delta(\omega) + \sigma_{s,\text{reg}}(\omega). \tag{17}
\]

We refer to Kubo formula for electrical conductivity \(^8\) and spin conductivity \(^2,3\) obtaining the regular part of spin conductivity \( \sigma_{s,\text{reg}}(\omega) \) for \( T = 0 \) in spectral representation

\[
\sigma_{s,\text{reg}}(\omega) = \frac{\pi \Omega}{N} \sum_{E_m \neq E_0} |\langle m | J_s(q) | 0 \rangle|^2 \delta(|\omega| - (E_m - E_0)) \frac{E_m - E_0}{E_m - E_0}. \tag{18}
\]

The Drude weight can be evaluated as follows:

\[
D_s = \frac{\pi a_0}{N\Omega} \left( -\hat{T} \right) - I_{\text{reg}}, \tag{19}
\]

\[
I_{\text{reg}} = \int_{-\infty}^{\infty} \sigma_{s,\text{reg}}(\omega) d\omega. \tag{20}
\]

The first term in eq.(19) is related to a sum of conductivity for all frequencies. We will discuss in detail in the next subsection.

3.3 Sum rule

In this subsection, we derive an f-sum rule for a spin current operator in this model following ref.[3][9]. First, we show continuity equation of both lattices in Fourier representations in the long wavelength limit

\[
iq J_s(-q) = \frac{1}{\Omega} \partial_t S_z^0(-q). \tag{21}
\]

Then we calculate the frequency integral of eq.(18) with use of eq.(21)

\[
iq N \int_{-\infty}^{\infty} \sigma_s(\omega) d\omega = \sum_m \frac{\langle 0 | J_s(q) | m \rangle \langle m | \partial_t S_z^0(-q) | 0 \rangle}{E_m - E_0} \frac{E_m - E_0}{(E_m - E_0)}.
\]

We get the following equations by applying Heisenberg equation of motion

\[
iq N \int \sigma_s(\omega) d\omega = \langle 0 | [J_s(q), S_z^0(-q)] | 0 \rangle. \tag{23}
\]

We obtain the left hand side of eq.(23) by calculating \( i [J_s(q), S_z^0(-q)] \) without any approximations using the laboratory frame

\[
i [J_s(q), S_z^0(-q)] = -iq \frac{a_0}{\Omega} \sum_l J_l \left( S^0_l S^0_{l+\hat{x}} + S^0_l S^0_{l+\hat{x}} \right)
\]

\[
= -iq \frac{a_0}{\Omega} \hat{T}. \tag{24}
\]
Here, $\hat{T}$ denotes $xy$ part of exchange interaction in the laboratory frame

$$\hat{T} = \sum_l J (S_{x0}^l S_{y0}^{l+x} + S_{y0}^l S_{x0}^{l+x}).$$  \hspace{1cm} (25)$$

We see that spin conductivity in both lattices satisfies the f-sum rule by the preceding procedure

$$\int_{-\infty}^{\infty} \sigma_s(\omega) d\omega = \frac{\pi a_0}{N \Omega} \langle -\hat{T} \rangle. $$  \hspace{1cm} (26)$$

This is the exact forms of spin conductivity f-sum rule valid for any lattice form.

Next, we examine the left hand side of eq.(26), which is denoted as $I$, and classify various terms in $I$ coming from the Holstein-Primakoff expansion according to the powers of $S$

$$I = \sum_n I_n \quad n = 2, 1, 0 \ldots,$$  \hspace{1cm} (27)$$

where $I_n$ is a term proportional to $S^n$. We focus on

$$\langle \hat{T} \rangle = \sum_n \langle \hat{T}_n \rangle \quad n = 2, 1, 0 \ldots,$$  \hspace{1cm} (28)$$

where $\hat{T}_n$ is a term proportional to $S^n$, to derive the left hand side of eq.(26) in Holstein-Primakoff expansion smoothly. We don’t have to consider $\hat{T}_n/2$ when $n$ is odd integer, because their expectation values are always zero. We obtain $I_n$ by using the following equation

$$I_n = \frac{\pi a_0}{N \Omega} \langle -\hat{T}_n \rangle \quad n = 2, 1, 0 \ldots.$$  \hspace{1cm} (29)$$

We now examine the following $\hat{T}_2$ and $\hat{T}_1$, which are the first and second terms in Holstein-Primakoff expansion, for both lattices. For square lattice

$$\hat{T}_2 = -JS^2 \cos^2 \theta N$$  \hspace{1cm} (30)$$

$$\hat{T}_1 = 2JS \cos^2 \theta \sum_l n_l - JS^2 \left( \cos^2 \theta' - \cos^2 \theta \right) N$$

$$\quad + \frac{JS}{2} \sum_l \left( \sin^2 \theta + 1 \right) \left( a_i^\dagger a_{l+x} + a_{l+x}^\dagger a_i \right)$$

$$\quad + \frac{JS}{2} \sum_l \left( \sin^2 \theta - 1 \right) \left( a_i^\dagger a_{l+x}^\dagger + a_l a_{l+x} \right).$$  \hspace{1cm} (31)$$

Here $\cos^2 \theta'$ is obtained by considering quantum correction to the canting angle$^5$

$$\cos^2 \theta' = 1 - \sin^2 \theta \left( 1 + 2W \right),$$  \hspace{1cm} (32)$$

$$w = \frac{1}{N} \sum_k \left[ (1 - \gamma_k) v_k^2 - \gamma_k u_k v_k \right].$$  \hspace{1cm} (33)$$
For triangular lattice
\[
\hat{T}_2 = JS^2 \sum_l \sin \theta_{l_{x}} \sin \theta_l
\]
\[
\hat{T}_1 = \frac{JS}{2} \sum_l \left( \frac{\cos^2 \theta - 2 \cos \theta}{3} - 1 \right) \left( a_{l_{x}}^{\dagger} a_{l_{x}}^{\dagger} + a_l a_{l_x} \right)
\]
\[
+ \frac{JS}{2} \sum_l \left( \frac{\cos^2 \theta - 2 \cos \theta}{3} + 1 \right) \left( a_{l_{x}}^{\dagger} a_{l_{x}}^{\dagger} + a_l a_{l_x} \right)
\]
\[
- JS^2 \left( \frac{\sin^2 \theta' - \sin^2 \theta}{3} \right) N + JS \frac{2 \sin^2 \theta}{3} \sum_l n_l,
\]

here \( \cos^2 \theta' \) is obtained by the same procedure as square lattice
\[
\cos^2 \theta' = \frac{1}{4} \left( 1 - \frac{2h}{3JS} \left( 1 + \frac{w}{S} \right) + \left( \frac{h}{3JS} \right)^2 \left( 1 + 2 \frac{w}{S} \right) \right),
\]
and \( w \) is defined in eq.(33). We get the sum rule for the first and second terms in Holstein-Primakoff expansion by substituting \( \hat{T}_2 \) and \( \hat{T}_1 \) to eq.(29). We expect this formalism is valid and independent of lattice structures as far as magnetization is a conserved quantity.

4. Results and Discussions

In Fig. 1(a), we show the integrated intensity \( I \) of the spin conductivity, and corresponding quantity for the regular part \( I_{\text{reg}} \) at \( T = 0 \), to the leading order in Holstein-Primakoff expansion for square lattice. Here, \( I_{\text{reg}2} \) denotes leading contribution to \( I_{\text{reg}} \) in Holstein-Primakoff expansions. The leading term of spin conductivity is calculated by substituting \( J_s 3/2 \) for \( J_s \) in eq.(18) for each lattices. In Fig.1(a), we show the integrated intensities \( I_{\text{reg}2} \) and \( I_2 \), defined in eq.(20) and eq.(29), respectively. We intentionally shifted the curve for \( I_2 \) by small amount because two results overlap completely. We thus find that the Drude weight vanishes for square lattice at \( T = 0 \) for any magnetic field \( h \) by comparing Fig.1(a) to eq.(19), because the difference between these two results defines the Drude weight. The vanishing Drude weight at \( T = 0 \) is consistent with ref..\(^3,4\)

We compare the intensities \( I_2 \) to \( I_2 + I_1 \) in Fig.1(b). This figure indicates that there exists two kinds of magnetic-field effect on the corrected intensity. One is dominant at low fields and the other is dominant at high fields. Spin wave corrections on staggered magnetization due to zero point fluctuation suppress integrated intensity and its effect is dominant at low fields with small gap excitation at Γ point. Canting angle changes and saturates with increasing fields, locking spins toward field direction, thus suppressing the spin conductivity. This effect is dominant at high fields. Whereas the leading term \( I_2 \) monotonically decrease with magnetic
Fig. 1. (a) The leading term $I_2$ of the integrated intensity of the spin conductivity and corresponding quantity $I_{\text{reg} 2}$ for the regular part as functions of magnetic field on square lattice. We intentionally shift the curve for figures of $I_2$ because of the degeneracy of two results, which indicates vanishing of the Drude weight for any field at $T = 0$. (b) We compare $I_2$ and $I_2 + I_1$ as a function of a magnetic field on square lattice. $I_2$ monotonically decrease as a result of locking spins with increasing fields. Spin wave corrections on staggered magnetization strongly suppress $I_2 + I_1$ at low fields.

field $h$, the quantum corrected intensity $I_2 + I_1$ is now a non-monotonic function with two kinds of effects. We get similar results in triangular lattice which is not shown in this letter.

We show the frequency dependence of spin conductivity for square lattice in Fig. 2(a) and for triangular lattice in Fig. 2(b) both calculated to the leading order only. We see van-Hove singularity in each lattice for any static homogeneous magnetic field. We expect these singularities to be removed by considering $1/S$ corrections of Holstein-Primakoff expansions.3

We notice that the lowest and the highest values of excitation spectrum in magnetic Brillouin zone determine the threshold of low and high frequency limit of spin conductivity. We note that the lower bound of spin conductivity spectrum at low fields $h/J \leq 1$ is determined by the excitation gap at $\Gamma$ point, which is the lowest energy for both lattice as seen from eq.(8) and eq.(9). On the other hand, the $\Gamma$ point gap determines the upper bound of conductivity spectrum for sufficiently high magnetic fields, as shown Fig.2. We expect similar behavior in triangular lattice in higher magnetic field regions. These results indicate that the excitation spectrum of magnetic Brillouin zone essentially determines spin conductivity.
Fig. 2. (a) Results of the leading order of spin conductivity for square lattice. (b) Results of the leading order of spin conductivity for triangular lattice. These results of both lattices show van-Hove singularities and indicate shapes of the excitation spectrum affect results of the spin conductivity.

5. Conclusion

We have derived the spin current operator for Heisenberg antiferromagnets and the spin conductivity in square and triangular lattices. We have shown that the Drude weight vanishes at $T = 0$ for any external static magnetic field for square lattice within linear spin wave theory, which is consistent with ref. $^3, ^4$ Two kinds of magnetic-field effect on integrated intensities in spin conductivity are found and the competition between the two makes corrected intensity a non-monotonic function of the magnetic field $h$.

We have calculated the frequency dependence of spin conductivity for both lattices, which indicates that excitation spectrum of magnetic Brillouin zone determines spin conductivity. We expect to get more realistic results by taking $1/S$ corrections of Holstein-Primakoff expansions into account. Lastly, we believe this method is applicable to any value of $S \geq 1/2$ and any non-collinear as well as collinear antiferromagnet as far as magnetization is conserved.
References

1) F. Heidrich-Meisner, A. Honecker, and W. Brenig, Phys. Rev. B 71, 184415 (2005).

2) S. Langer et al., Phys. Rev. B 82, 104424 (2010).

3) M. Sentef, M. Kollar, and A. P. Kampf, Phys. Rev. B 75, 214403 (2007).

4) Z. Chen, T. Datta, and Dao-Xin Yao, Eur. Phys. J. B, 86: 63 (2013); A. S. T. Pires and L. S. Lima, Phys. Rev. B 79, 064401 (2009).

5) M. Mourigal, M. E. Zhitomirsky, and A. L. Chernyshev, Phys. Rev. B 82, 144402 (2010); M. E. Zhitomirsky and T. Nikuni, Phys. Rev. B 57, 5013 (1998).

6) A. L. Chernyshev and M. E. Zhitomirsky, Phys. Rev. B 79, 144416 (2009).

7) H. Kawamura, J. Phys. Soc. Jpn. 53, 2452 (1984).

8) G. D. Mahan: Many-Particle Physics (Plenum Press, New York, 2000) 3rd ed., p. 160.

9) P. F. Maldague, Phys. Rev. B 16, 2437 (1977).