Magnon Pairs and Spin-Nematic Correlation in the Spin-Seebeck Effect

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Investigating exotic magnetic materials with spintronic techniques is effective at advancing magnetism as well as spintronics. In this work, we report unusual field-induced suppression of the spin-Seebeck effect (SSE) in a quasi one-dimensional frustrated spin-1/2 magnet LiCuVO4, known to exhibit spin-nematic correlation in a wide range of external magnetic field B. The suppression takes place above |B| > 2 T in spite of the B-linear isothermal magnetization curves in the same B range. The result can be attributed to the growth of the spin-nematic correlation while increasing B. The correlation stabilizes magnon pairs carrying spin-2, thereby suppressing the interfacial spin injection of SSE by preventing the spin-1 exchange between single magnons and conduction electrons at the interface. This interpretation is supported by integrating thermodynamic measurements and theoretical analysis on the SSE.

Introduction—Spin-Seebeck effects (SSE)1–19 refer to the generation of a spin current owing to a temperature gradient in a magnetic material. It takes place in a magnetic insulator with a metallic contact. When nonequilibrium magnons are accumulated at the interface due to a temperature gradient, the annihilation of such a single magnon is followed by the flip of a conduction-electron spin via the interfacial exchange interaction. As a result, the exchange of spin-1 takes place dominantly, enabling conversion from a magnon spin current into a conduction-electron one [3]. The latter spin current can be detected as a transverse electric field via the inverse spin-Hall effect [20, 23] in the metallic contact. SSEs have been found to take place even in paramagnetlike insulators with spin correlations [16, 18]. These findings point to the use of SSE as a probe for spin correlations without the magnetic orders, for example, in quantum spin systems [24, 27].

The magnetic quadpolar correlation, also known as the spin-nematic correlation [25–30], is the simplest example of magnetic multipolar correlations. It represents the correlation between magnon pairs, rather than single magnons. To stress this point, the spin-nematic correlation will be called the magnon-pair correlation hereafter. A typical magnon-pair correlation appears in a one-dimensional (1D) frustrated spin-1/2 chain with the ferromagnetic nearest neighboring exchange interaction J1 < 0 and the antiferromagnetic next nearest neighboring one J2 > 0. The Hamiltonian of this 1D J1-J2 model reads

$$\mathcal{H} = \sum_{j} (J_{1} S_{j} \cdot S_{j+1} + J_{2} S_{j} \cdot S_{j+2} - g \mu B B S_{j}^z).$$

(1)

Here Sj is the spin-1/2 operator on the jth site, and the site number j increases along the spin-chain direction. The last term represents the Zeeman interaction with the external magnetic field B along the z-axis with g and \(\mu_B\) being respectively the g-factor and the Bohr magneton. The low-energy physical properties of Eq. (1) and its variants have been elucidated [31, 32] using powerful theoretical techniques in the last decade. The ground-state diagram with \(|J_{1}/J_{2}| = O(1)\) [32, 33] is schematically shown in Fig. 1(a) as a function of B. In the lower B range, a Tomonaga-Luttinger liquid (TLL) [27] with a vector spin chirality [34, 35] appears. As B is increased, the magnon-pair correlation grows to give rise to a spin-nematic TLL [31, 36] in a wide B range. In this state, single magnons acquire an energy gap equivalent to the binding energy of magnon pairs while magnon pairs are gapless. Accordingly, a change in spin angular momentum is quantized in units of 2h, not \(\hbar\), in low energy.

In this study, we have investigated the SSE in an insulating quantum magnet LiCuVO4 [32, 35]. LiCuVO4 is an established model material for a strong magnon-pair correlation, representing a family of quasi-1D J1-J2 magnets [38, 50]. Since the spin quantum number carried by quasiparticles is increased effectively by magnon-pair formation, the SSE seems to be enhanced while increasing B. Contrary to this naïve expectation, the SSE in LiCuVO4 has been observed to exhibit a strong B-induced suppression alongside the B-linear magnetization curves above the magnetic ordering temperatures. Such a B response of the SSE is different from those of magnetically ordered states [11, 12, 14] and a 1D quantum spin liquid [16]. We interpret the result as the evidence for B-induced crossover from the single-magnon correla-
tion to the magnon-pair one, and its resulting prevention of the interfacial exchange of spin-1 in the SSE. Observing the magnon-pair correlation is generally difficult. Our study shows that SSE serves as a powerful probe for dynamical and transport natures of such spin-nematic states in quantum magnets.

**Spin-nematic nature of LiCuVO$_4$—** LiCuVO$_4$ is a typical Mott insulator for which experimental evidences for the magnon-pair correlation have been established. A spin chain embedded in LiCuVO$_4$ is shown in Fig. 1(b). Each Cu$^{2+}$ ion carries spin-$\frac{1}{2}$ and they form a 1D chain along the $b$-axis by sharing O$^{2-}$ ions. If the weak interchain interaction $J'$ is ignored, LiCuVO$_4$ can be well described by Eq. (1). The magnitudes of $J_1$ and $J_2$ were estimated experimentally, for example, from neutron scattering spectra [42, 44, 46, 47]: $J_2 = 40 \sim 70$ K and $|J_1/J_2| = O(1)$. Because of the weak $J'$ in LiCuVO$_4$, magnetically ordered phases appear at low temperatures; however, each phase nicely reflects the phase diagram of magnetically ordered phases reported elsewhere [45], as shown in Fig. 1(c). The experimental details and the magnetic properties are described in Supplemental Material [64].

We used a LiCuVO$_4$/Pt junction system as shown in Fig. 1(d) to investigate the SSE. A temperature gradient $\nabla T$ was applied along the spin chains with a heater. We created the temperature difference $\Delta T$ between the top of the Pt film and the rear of the LiCuVO$_4$. Au wires were attached to the ends of the Pt film to obtain the DC voltage $V$, for which we excluded a background voltage signal taken with the heater off. The magnetic field $B$ was applied along the $c$-axis, being perpendicular both to $\nabla T$ and the direction across the electrodes; thus, the $c$-axis corresponds to the $z$-axis in Eq. (1) while the $a$- and $b$-axes to the $x$- and $y$-axes, respectively. To quantitatively compare the voltage signals, we show the transverse thermopower $S = j_e/|\nabla T| \approx (V/\Delta T \rho)(t/l)$. Here $j_e$ is the current density in the Pt film due to thermoelectric effects, $\rho$ is the electrical resistivity of the Pt film, and $t$ and $l$ are respectively the thickness and the length of the LiCuVO$_4$. Additionally, we defined the average temperature $T_{ave}$ as $T_{ave} = (T_H + T_L)/2$ in which $T_H = T + \Delta T$ and $T_L = T$ are respectively the temperatures of the top of the Pt film and the rear of the LiCuVO$_4$. The experimental details of SSE measurements are described in Supplemental Material [64].

**Experimental details—** Single crystals of LiCuVO$_4$ were grown by a travelling-solvent floating-zone method, which was exactly the same as reported one of the present authors [62]. The grown single crystals were cut into cuboids that were typically 5 mm along the $a$-axis and 1 mm along the $b$- and $c$-axes for SSE measurements. Temperature ($T$) and magnetic field ($B$) dependences of the magnetization were found to be consistent with a $B-T$ phase diagram reported elsewhere [45], as shown in Fig. 1(c). The experimental details and the magnetic properties are described in Supplemental Material [64].

**FIG. 1:** (a) Theoretical ground-state phase diagrams of a purely 1D frustrated $J_1$-$J_2$ spin-$\frac{1}{2}$ chain [32, 33] (top) and a quasi-1D one with an interchain exchange interaction [36] (bottom). $B$ denotes external magnetic field. (b) Spin chain in LiCuVO$_4$ composed of Cu$^{2+}$ and O$^{2-}$ ions. (c) Magnetic field ($B$) – temperature ($T$) phase diagram of LiCuVO$_4$, obtained while applying $B$ in the $c$-axis. Triangular data points were taken in this study: the sky-blue ones from the $T$ dependence of the magnetization $M$; the orange ones from the $B$ dependence of $M$. The circular data points were adapted from Ref. [45] (d) Experimental set-up for detecting the spin-Seebeck effect in a LiCuVO$_4$/Pt system. $J_1$ and $J_2$ respectively denote the nearest and next nearest neighboring exchange interactions in the spin chain of LiCuVO$_4$; $\nabla T$ a temperature gradient along the spin chain; $t$ and $l$ respectively the thickness and the length of the LiCuVO$_4$.
increased down to 11 K, a clear signal appears. Its sign
reverses when the magnetization is reversed, which is a
typical feature of SSE. Interestingly, $S$ starts deviating
from a $B$-linear line, and decreases while increasing $B$.
As shown in Fig. 2(b), the deviation enhances with a fur-
ther decrease of $T_{\text{ave}}$ down to 2 K, the lowest temperature
in this study.

To look into this $B$-dependence of $S$ in more detail, we
compare the $B$ dependences of $S$ and the magnetization
$M$ at $T = 4$ K in Fig. 2(c). Remarkably, in spite of the
$B$-linear change in $M$, $S$ gets suppressed strongly
while increasing $B$, and even exhibits a negative slope at
$|B| > 5$ T. We stress that the suppression of $S$ cannot
be attributed to magnetic phase transitions since it takes
place even above $T_{N}$ [see also Fig. 1(c)]. Additionally, the
Zeeman energy gap in spin excitations is unlikely to ex-
plain the $B$-induced suppression of $S$ although seemingly
similar results were reported for ferrimagnets and param-
agnets. $B$-linear. Generally the Zeeman energy gap starts
suppressing thermal magnetic excitations as the magneti-
zation approaches saturation at low temperatures. Since
$M$ of LiCuVO$_4$ is $B$-linear alongside $\sim 0.1$ $\mu_B$/Cu$^{2+}$
even at $B = 9$ T, the smooth $M - B$ curve indicates the exis-
tence of a gapless magnetic excitation.

The unusual suppression of $S$ invokes the magnon-pair
correlation, which yields magnon pairs with a binding
energy $E_{\text{bind}}$. $E_{\text{bind}}$ has been predicted to already ex-
ist near zero magnetic field. $S$ exhibits a $B$ dependence of $E_{\text{bind}}$ for a purely 1D case with $|J_1/J_2| = 1$. $E_{\text{bind}}$ increases linearly with $B$ alongside the $B$-linear magnetization when $B$ is much lower than the saturation field [see also the inset to Fig. 3(a)]. Within this framework, the $B$-induced $E_{\text{bind}}$ stabilizes magnon pairs while inhibiting thermal excitation of single magnons. Because spin injection of SSE at the interface stems mainly from the exchange of spin-
1, spin-2 magnon pairs cannot contribute to such spin
injection, thereby decreasing SSE signals. The ability to
selectively probe spin-1 magnetic excitations should differ-
entiate SSE measurements from thermal conductivity
measurements. This is because the latter measurements
simultaneously probe phonons as well as multiple mag-
netic excitations carrying spin-1 and spin-2.

**Comparison between experimental and theoretical results**— We theoretically calculate spin currents injected
from a magnet (LiCuVO$_4$) to a metal (Pt) and compare
them with $S$, because inverse spin-Hall voltages are pro-
portional to injected spin currents. For simplicity, we as-
sume that the spin dynamics of LiCuVO$_4$ is described by
a spin-nematic TLL, ignoring the weak inter-chain in-
teractions. We also make the conventional assumption
that a weak exchange interaction $J_{\text{ad}}$ exists at the interface
between the magnet and the metal. The normalized spin
current $\tilde{J}_s$ is then given by (see Supplemental Material)

$$\tilde{J}_s = \frac{1}{T^2} \int d\omega \Im \chi_{\text{mag}} (\omega, T) \frac{\omega^2}{1 + \tau_\omega^2 \omega^2} \frac{1}{\sinh^2 (\omega/(2T))}$$

up to the leading order of $J_{\text{ad}}$. Here, $\omega$ is the angular fre-
quency, $T$ is the mean value of the two temperatures of
the magnet and the metal, and $\tau_\omega$ is the spin relaxation
time for the metal. The integral range is $(-\infty, \infty)$. $\chi_{\text{mag}}$
denotes the dynamical spin susceptibility of the magnet,
and describes the dynamics of a single magnon (strictly
speaking, a paramagnon in a spin-nematic TLL). In for-
mula (2), the spin current is injected by single magnons
which have an energy gap due to magnon-pair formation.
We have ignored the magnon-pair-driven spin cur-
rent considering its small magnitude. Magnon-pair
formation is considered via the resulting energy gap in
$\chi_{\text{mag}}$, whose low-energy form at finite temperatures was
determined within the framework of practical approxi-
mation.

In Fig. 3(b), we show the $B$ dependences of calculated
$\tilde{J}_s$ and measured $S$ at $T = 4$ K normalized by their max-
imum values. We set $J_1/J_2 = -1$ and $J_2 = 50$ K in
the calculation and normalized $B$ by the saturation field
$B_s$ (see also the caption of Fig. 3). $\tilde{J}_s$ and $S$ increase
linearly with $B$ near zero magnetic field. This can be
attributed to the growth of the uniform ferromagnetic
moment and the angular momentum along $B$ per sin-
FIG. 2: (a), (b) $B$ dependence of the transverse thermopower $S$ at several $T$. (c) Comparison between $B$ dependences of $S$ and $M$ at $T = 4$ K.
The low-B SSE is similar to antiferromagnetic SSEs in canted phases \[12\, 13\]. In these previous cases, spin-Seebeck coefficients exhibit positive sign along with the same B dependences as those of M. These features are expected to be embedded in our low-B SSE results. Integrating such effects into the above calculation will yield a more quantitative result while the B-induced suppression of magnon-pair origin should carry over.

In Fig. 4, we compare the \( T_{\text{ave}} \) dependences of \( S \) for several \( B \) with our theoretical calculations, in which finite-temperature effects on the single-magnon dynamics are considered besides the magnon-pair binding energy. When \( B \) is below \( \sim 5 \) T, \( S \) only saturates toward low \( T_{\text{ave}} \) as seen in Fig. 4(a). However, when \( B \) is above \( \sim 5 \) T, a broad peak structure emerges, and its peak position gradually shifts from \( \sim 5 \) K to \( \sim 8 \) K while increasing \( B \) to 14 T. These temperature dependences are also successfully captured by our calculation based on formula (2), as shown in Fig. 4(b). This shows that the broad peaks stem from the competition between a decrease in the single-magnon density due to the magnon-pair formation and an increase in the single-magnon lifetime at low temperatures. Additionally, the agreement between Figs. 4(a) and (b) indicates that the peak shift caused by increasing \( B \) could be attributed to an increase in the angular momentum along \( B \) per single magnon [39]. Such increased angular momentum enhances SSE at high temperature where the \( B \)-induced magnon-pair binding energy can be overcome by thermal fluctuation; otherwise, SSE is decreased more greatly toward low temperature via magnon-pair formation.

This can be responsible for the peak shift observed in Fig. 4(a). Overall, the agreement between the experimental
gle magnon [39]. Most importantly, \( \dot{J}_s \) starts to be suppressed upon a further increase of \( B \), and exhibits a broad peak structure around \( |B| = 9 \) T, capturing the marked feature of \( S \) observed experimentally. Since applying \( B \) of this magnitude yields \( E_{\text{bind}} \sim 3 \) K [see also Fig. 3(a)], the \( B \)-induced suppression at \( T = 4 \) K can be ascribed to a decrease in thermally excited single magnons that is induced by magnon-pair formation. We stress that the theoretical \( B_s \) is varied easily by changing \( J_1 \) and \( J_2 \) [32] in the spin-nematic TLL state while the \( B \)-linearity of \( E_{\text{bind}} \) is not [39]. Thus, the \( B \) dependence of \( \dot{J}_s \) little depends on change of \( B_s \). This indicates that a difference between the theoretical \( B_s = 93 \) T and the experimental \( B_s \sim 43 \) T is not essential in reproducing the characteristic \( B \)-dependence of \( S \).

We note that for LiCuVO\(_4\), the 3D spin spiral correlation likely coexists with the magnon-pair one above magnetic ordering temperatures \( \sim 3 \) K [see also Fig. 1(c)]. Since \( B \) is applied parallel to the spiral axis along the c-axis, the low-B SSE is similar to antiferromagnetic SSEs in canted phases \[12\, 13\]. In these previous cases, spin-seebeck coefficients exhibit positive sign along with the same \( B \) dependences as those of \( M \). These features are expected to be embedded in our low-B SSE results. Integrating such effects into the above calculation will yield a more quantitative result while the \( B \)-induced suppression of magnon-pair origin should carry over.
and theoretical results shows that the $B$ and $T$ dependences of $S$ can be well explained by magnon-pair formation. We also note that our results point to exchange of spin-1 as the most relevant magnetic interaction at the interface in SSE.

Summary— We observed the magnetic-field-induced suppression of the SSE in a quasi-1D frustrated spin-chain system LiCuVO$_4$, an established model material for the spin-nematic correlation. A broad peak structure was also found to appear in the temperature dependence of the spin-Seebeck voltage, and to shift toward high temperatures while increasing magnetic field. These experimental results were well reproduced by a microscopic calculation of the interfacial spin current where the magnon-pair binding energy and its resulting energy gap of the single magnons are taken into consideration. Our result indicates that SSE is a powerful tool for detecting signatures of spin-nematic states and their transport properties.

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