Magnetothermal transport in the spin-$\frac{1}{2}$ easy-axis antiferromagnetic chain

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By an exact analytical approach we study the magnetothermal transport in the spin-$\frac{1}{2}$ easy-axis Heisenberg model, in particular the thermal conductivity and spin Seebeck effect as a function of anisotropy, magnetic field, and temperature. We stress a distinction between the common spin Seebeck effect with fixed boundary conditions and the one (intrinsic) with open boundary conditions. In the open boundary spin Seebeck effect we find exceptional features at the critical fields between the low field antiferromagnetic phase, the gapless one, and the ferromagnetic at high fields. We further study the development of these features as a function of easy-axis anisotropy and temperature. We point out the potential of these results to experimental studies in spin chain compounds—candidates for spin current generation in the field of spintronics.

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I. INTRODUCTION

Over the past couple of decades the magnetic thermal transport has been established as a very efficient mode of thermal conduction [1], next to the well known phononic and electronic ones. Parallel to the search for evidence of ballistic thermal transport in quasi-one-dimensional spin chain compounds described by the spin-$\frac{1}{2}$ Heisenberg Hamiltonian [2,3], numerous experimental studies focused on the effect of a magnetic field on the thermal conductivity. For instance, the spin-$\frac{1}{2}$ copper pyrazine dinitrate [4], spin-one NENP [5], and ladder compounds [6] were experimentally studied focusing on the interplay between the contributions of magnetic and phononic excitations and their mutual scattering. Besides the magnetic thermal transport, only few recent studies were devoted to the spin Seebeck effect in quantum spin liquid systems, namely the generation of a spin current by a thermal gradient in a magnetic field—for instance, experimental studies on the Sr_2CoO_3 [7] compound with topological spinon excitations, $CuGeO_3$ with triplon excitations [8], the spin-$\frac{1}{2}$ easy-axis antiferromagnet $Pb_2V_3O_9$ [9], and theoretical ones [10–13].

From a different perspective, the generation and control of spin currents is a central topic in the field of spintronics [14]. In particular, the spin Seebeck effect [15] has been extensively experimentally and theoretically studied in a great variety of bulk magnetic compounds such as the ferrimagnetic YIG/Pt heterostructures and antiferromagnetic materials, e.g., Cr_2O_3 and Fe_2O_3. Concerning the easy-axis antiferromagnetic materials, there is experimental and theoretical interest and debate on the generated spin current sign change at the spin-flop transition.

Motivated by the abundance of Ising-like antiferromagnetic spin chain compounds and experimental studies over the years, e.g., on $CsCoCl_3$, $CsCoBr_3$, TMMC in the context of soliton Villain excitations [16], phase diagram, spin dynamics, and quantum criticality of $ACo_2V_2O_8$ (A = Sr, Ba, Pb) [17–19], we study the magnetothermal transport and in particular the spin Seebeck coefficient in the spin-$\frac{1}{2}$ easy-axis antiferromagnetic Heisenberg model. This study also serves as a bridge between spintronics studies in bulk materials and prototype magnetic systems. We employ the thermodynamic Bethe ansatz (TBA) approach to analytically evaluate the relevant spin-energy current correlations within linear response theory. We explore, in particular, the sign of the spin current across the antiferromagnetic, gapless, and ferromagnetic phases that characterize the Ising-like antiferromagnetic Heisenberg chain and the singular behavior at the critical fields.

II. MODEL AND METHOD

We study the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg model with easy-axis anisotropy, given by the Hamiltonian

$$H = \sum_{l=1}^{L} J_{\perp}\left(S_l^x S_{l+1}^x + S_l^y S_{l+1}^y\right) + \Delta S_l^z S_{l+1}^z - h S_l^z, \qquad (1)$$

where $S_l^{x,y,z} = \frac{1}{2}\sigma^{x,y,z}$, $\sigma^{x,y,z}$ are Pauli spin matrices, and $J_{\perp} > 0$ is the easy plane and $\Delta > 0$ the easy-axis exchange interactions with $\Delta > J_{\perp}$ and h the magnetic field. Hereafter, we take $J_{\perp} = 1$ as the unit of energy.

In linear response theory the spin and energy currents are related by the transport coefficients, C_{ij},

$$\begin{pmatrix} \langle J^Q \rangle \\ \langle J^S \rangle \end{pmatrix} = \begin{pmatrix} C_{QQ} & C_{QS} \\ C_{SQ} & C_{SS} \end{pmatrix} \begin{pmatrix} -\nabla T \\ \nabla h \end{pmatrix}, \qquad (2)$$

where $C_{QQ} = \kappa_{QQ}$ ($C_{SS} = \sigma_{SS}$) is the heat (spin) conductivity and the thermal current J^Q is related to the energy J^E and spin current J^S by $J^Q = J^E - hJ^S$. The coefficients C_{ij} are given by the thermal average of time-dependent current-current correlation functions and it is straightforward to see that $C_{SQ} = \beta C_{QS}$ ($\beta = 1/k_BT, k_B = 1$). The real part of $C_{ij}(\omega)$ can be decomposed into a δ function at $\omega = 0$ (the Drude weight) and a regular part:

$$\mathrm{Re}[C_{ij}(\omega)] = 2\pi D_{ij}\delta(\omega) + C_{ij}^{\mathrm{reg}}(\omega). \tag{3}$$

The spin-$\frac{1}{2}$ Heisenberg model is integrable by the Bethe ansatz method and transport is ballistic at finite magnetic fields [3], with the energy current commuting with the Hamiltonian. Thus the magnetothermal coefficients are given by the Drude weights D_{ij}. We should note that, in view of experiments, this correspondence holds only if we assume the same relaxation rates for the magnetization and energy transport, $C_{ij} \sim D_{ij}\tau$ [10–13]. We will consider the following situations.

(i) $\langle J^S\rangle = 0$, corresponding to a "fixed boundary" system with spin accumulation, giving the thermal conductivity κ,

$$\kappa = D_{QQ} - \beta\frac{D_{QS}^2}{D_{SS}} = D_{EE} - \beta\frac{D_{ES}^2}{D_{SS}}. \tag{4}$$

$\mathrm{MTC} = \beta\frac{D_{QS}^2}{D_{SS}}$ is the magnetothermal contribution and the spin Seebeck coefficient,

$$S = \frac{\nabla h}{\nabla T} = \beta\left(\frac{D_{ES}}{D_{SS}} - h\right). \tag{5}$$

(ii) $\nabla h = 0$, corresponding to an "open boundary" system [20], also referred to as intrinsic or bulk spin Seebeck effect [21],

$$\tilde{S} = \frac{\langle J^S\rangle}{\langle J^Q\rangle} = \beta\frac{D_{QS}}{D_{QQ}}, \tag{6}$$

where $D_{QQ} = D_{EE} - 2\beta hD_{ES} + \beta h^2D_{SS}$ and $D_{QS} = D_{ES} - hD_{SS}$.

We evaluate the magnetothermal Drude weights in the framework of the TBA [22–26] approach. In the easy-axis regime [22] the anisotropy is parametrized as $\Delta = \cosh\eta$ and in contrast to the easy-plane regime the Bethe ansatz solution is characterized by an infinite number of string excitations with "particle" (and "hole") densities $\rho_j(x)[\rho_j^h(x)]$, $j = 1, \infty$, x pseudomomenta. D_{SS}, D_{ES}, D_{EE}, the specific heat C, and magnetic susceptibility are now given by the fairly standard TBA expressions

$$\begin{aligned}
D_{SS} &= \beta\sum_j\int_{-\pi}^{+\pi} dx\, r_jn_j(1-n_j)\big(v_j^QQ_j\big)^2,\\
D_{ES} &= \beta\sum_j\int_{-\pi}^{+\pi} dx\, r_jn_j(1-n_j)\big(v_j^EE_j\big)\big(v_j^QQ_j\big),\\
D_{EE} &= \beta^2\sum_j\int_{-\pi}^{+\pi} dx\, r_jn_j(1-n_j)\big(v_j^EE_j\big)^2,\\
C &= \beta^2\sum_j\int_{-\pi}^{+\pi} dx\, r_jn_j(1-n_j)(E_j)^2,\\
\chi &= \beta\sum_j\int_{-\pi}^{+\pi} dx\, r_jn_j(1-n_j)(Q_j)^2.
\end{aligned} \tag{7}$$

The total densities $r_j = \rho_j + \rho_j^h$ are obtained from

$$r_j = \big(\rho_j + \rho_j^h\big) = \frac{1}{\pi}a_j - \sum_k T_{jk} * \rho_k, \tag{8}$$

where "$*$" denotes convolution symbol

$$\begin{aligned}
a_j(x) &= \frac{\sinh(j\eta)}{\cosh(j\eta) - \cos(x)},\\
T_{jk} &= (1-\delta_{jk})a_{|j-k|} + 2a_{|j-k|+2}\\
&\quad + \cdots + 2a_{j+k-2} + a_{j+k},\\
f^*g(x) &= \frac{1}{2\pi}\int_{-\pi}^{+\pi} f(x-y)g(y)dy,
\end{aligned}$$

and the occupation numbers $n_j = 1/(1+e^{\beta\epsilon_j})$, from the thermal energies ϵ_j,

$$\epsilon_j = \epsilon_j^{(0)} + T\sum_k T_{jk} * \ln(1+e^{-\beta\epsilon_k}), \tag{9}$$

where $\epsilon_j^{(0)} = -\sinh\eta\cdot a_j(x) + hj$ are the bare excitation energies. The effective velocities are given by [28]

$$v_j^E = -v_j^Q = \frac{1}{2\pi r_j}\frac{\partial\epsilon_j}{\partial x}, \tag{10}$$

the "dressed" charges Q_j and energies E_j,

$$\begin{aligned}
Q_j &= Q_j^{(0)} - \sum_k T_{jk} * (n_kQ_k), \quad Q_j^{(0)} = j,\\
E_j &= \epsilon_j^{(0)} - \sum_k T_{jk} * (n_kE_k),
\end{aligned} \tag{11}$$

and the magnetization,

$$\langle S^z\rangle = \frac{1}{2} - \frac{1}{2}\int_{-\pi}^{+\pi} dx\, r_jn_jQ_j. \tag{12}$$

In the $T \to 0$ limit, there are three different phases [27,28]: (i) for $h < h_c = \sqrt{\Delta^2-1}\cdot\mathrm{Dn}(\pi)$ it is gapped antiferromagnetic, (ii) for $h_c < h < h_f = 1+\Delta$ it is a gapless spin liquid, and (iii) for $h > h_f$ it is gapped ferromagnetic [$\mathrm{Dn}(x) = \frac{1}{2}\sum_{j=-\infty}^{+\infty}\frac{e^{ijx}}{\cosh(jx)}$ is the elliptic Jacobi function].

As $\epsilon_1 < 0$, $\epsilon_j > 0$ for $j > 1$ in the low field antiferromagnetic phase, we find that Eqs. (9) and (11) solved numerically by iteration with a finite cutoff in the number of strings show poor or no convergence. The same applies when the thermal energies ϵ_j are numerically evaluated by the formulation of Ref. [22] and furthermore the evaluated quantities do not accurately satisfy "dressing" relations, as for instance Eqs. (8) and (11) should imply $\int dx\, r_jn_jQ_j^{(0)} = \int dx\, a_jn_jQ_j$.

To resolve the convergence and "dressing" issues, we transform and solve by iteration Eqs. (9) by rewriting the term $\ln(1+e^{-\beta\epsilon_1}) = -\beta\epsilon_1 + \ln(1+e^{\beta\epsilon_1})$ [29]. Fourier

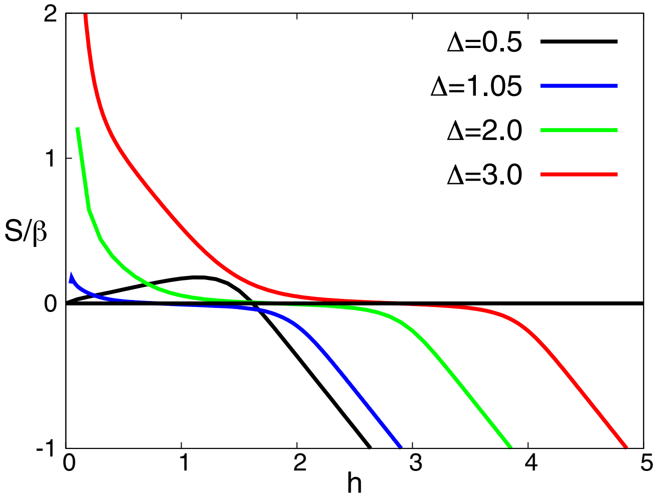

FIG. 1. Evolution of the spin Seebeck coefficient with anisotropy Δ as a function of magnetic field at temperature $T = 0.1$.

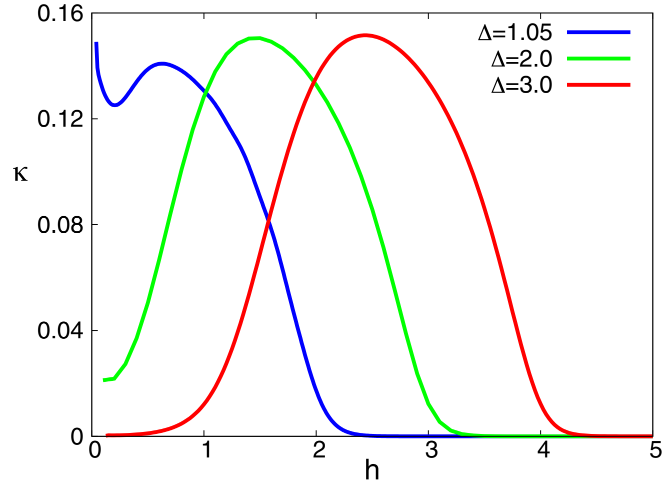

FIG. 2. Evolution of thermal conductivity with anisotropy Δ as a function of magnetic field at temperature $T = 0.1$.

transforming the equation for $j = 1$ ($\hat{\ldots}$ denotes Fourier transform),

$$\hat{\epsilon}_1 = \frac{\hat{\epsilon}_1^{(0)}}{1+\hat{T}_{11}} + T \cdot \frac{\hat{T}_{11}}{1+\hat{T}_{11}} \cdot \ln(1 \hat{+} e^{\beta\epsilon_1}) + T \cdot \sum_{k>1} \frac{\hat{T}_{1k}}{1+\hat{T}_{11}} \cdot \ln(1 \hat{+} e^{-\beta\epsilon_k}),$$

back transforming,

$$\tilde{\epsilon}_1 = \tilde{\epsilon}_1^{(0)} + T \cdot \tilde{T}_{11} * \ln(1+e^{\beta\epsilon_1}) + T \cdot \sum_{k>1} \tilde{T}_{1k} * \ln(1+e^{-\beta\epsilon_k}),$$

and repeating the substitution for $j > 1$, we obtain zero effective thermal energies $\tilde{\epsilon}_j^{(0)}$, densities $\tilde{r}_j^{(0)}$, and charges $\tilde{Q}_j^{(0)}$,

$$\begin{aligned} \tilde{\epsilon}_1^{(0)} &= -\sinh(\eta) \cdot \mathrm{Dn}(x) + \frac{h}{2}, \quad \tilde{\epsilon}_{j>1}^{(0)} = (j-1)h, \\ \tilde{r}_1^{(0)} &= \mathrm{Dn}(x), \quad \tilde{r}_{j>1}^{(0)} = 0, \\ \tilde{Q}_1^{(0)} &= \frac{1}{2}, \quad \tilde{Q}_{j>1}^{(0)} = j-1. \end{aligned} \tag{13}$$

Note that the obtained effective energies are identical to those obtained in the low temperature antiferromagnetic regime [27,28].

III. RESULTS

In Fig. 1 we show the spin Seebeck coefficient S at low temperature in the easy axis and for comparison for $\Delta = 0.5$ in the easy-plane regime. We find that S, in the gapless phase $h_c < h < h_f$, decreases with decreasing anisotropy, diverges as $h \to 0$, and changes sign between the antiferromagnetic and ferromagnetic phases. In contrast to the easy-plane regime where the spin Drude weight D_{SS} is finite and $S \to 0$ as $h \to 0$, the vanishing of D_{SS}, D_{ES} at $h = 0$ for $\Delta > 1$ implies an ill-defined S. The results we find are consistent with the spin Seebeck coefficient evaluated at the isotropic limit [11]. Of course, we expect physically the vanishing spin Drude weight D_{SS} at $h = 0$ to be replaced by a normal transport behavior, also at low temperatures, although this is still debated in studies focused in the high temperature limit [30].

In Fig. 2, the thermal conductivity κ is finite as $h \to 0$, although strongly suppressed in the gapped antiferromagnetic and ferromagnetic phase for large anisotropy Δ. In particular κ tends to a finite value as $h \to 0$ as the energy current commutes with the Hamiltonian and the thermal transport is purely ballistic over the whole phase diagram.

In Fig. 3 we show the ratio $\tilde{S}$ of the induced spin current to the thermal current. First, note that $\tilde{S}$ goes to zero as $h \to 0$ and there is a change of sign between the antiferromagnetic-gapless phase and the ferromagnetic one. However, pronounced features are developing at the critical fields h_c, h_f. It is remarkable that a similar behavior was found in a molecular dynamics and linear response study of the classical easy-axis Heisenberg model [20].

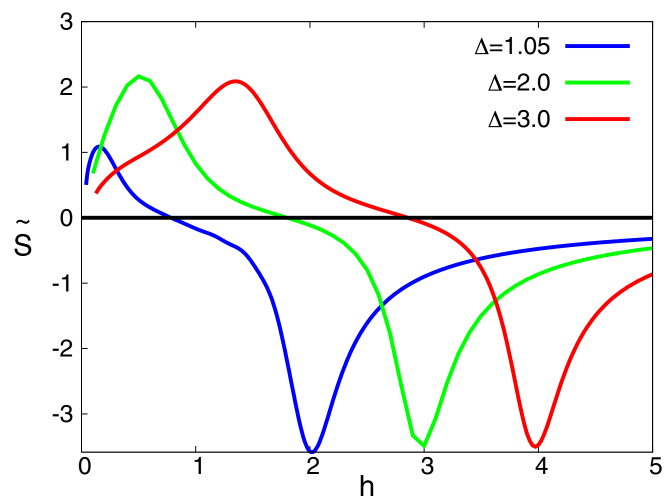

FIG. 3. Evolution of the ratio spin current to thermal current $\tilde{S}$ with anisotropy Δ as a function of magnetic field at temperature $T = 0.1$.

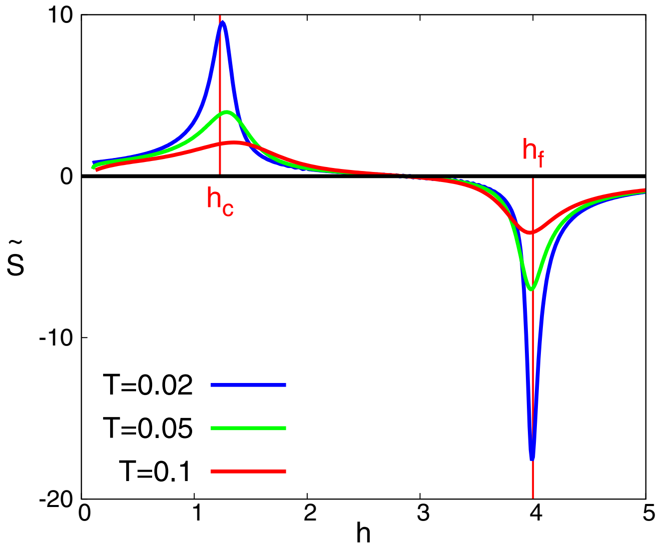

FIG. 4. Evolution of the ratio spin current to thermal current $\tilde{S}$ with temperature as a function of magnetic field at anisotropy $\Delta = 3$.

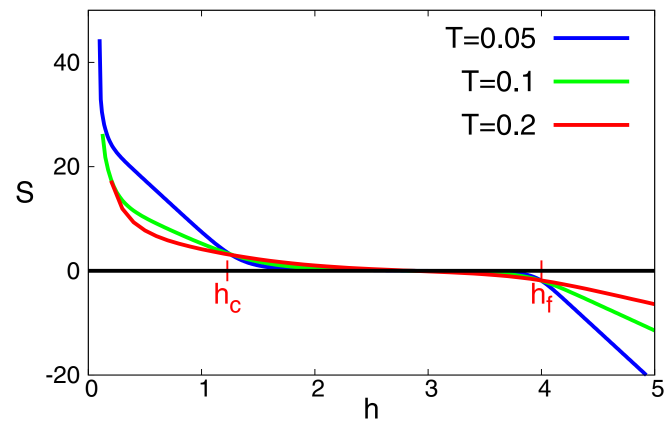

FIG. 6. Spin Seebeck coefficient as a function of magnetic field at different temperatures.

To further study the behavior at the phase transitions, in Fig. 4 we show $\tilde{S}$ lowering the temperature at a rather large anisotropy $\Delta = 3$ along with the critical fields h_c, h_f. In the antiferromagnetic phase, the spin current vanishes for $h \to 0$ in contrast to S. At the transition between the gapless and the ferromagnetic phase there is a particularly pronounced peak at the critical field h_f. This peak, as shown in Fig. 5, is related to the singular behavior of the specific heat and magnetic susceptibility at h_f [31]. It can be understood as the effect of the van Hove singularity in the density of states in the low density magnon system approaching the saturation field. The peaks at the critical fields h_c, h_f can be approximately described as Lorentzians of width proportional to the temperature.

In Fig. 5, we show the increase of magnetization as a function of magnetic field, from zero to saturation. Note that there is no exceptional singular behavior of the thermodynamic quantities at h_c, although there is one in $\tilde{S}$ as shown in Fig. 4. We should note that the singularities in $\tilde{S}$ are related to the numerator D_{QS} in Eq. (6), which follows a very similar pattern as a function of magnetic field (not shown).

Figure 6 shows the spin Seebeck coefficient S at different temperatures. It indicates a vanishing S in the gapless phase as the temperature tends to zero, in accord with the induced spin current shown in Fig. 4 and with calculation [13] in the gapless easy-plane ($\Delta < 1$) regime. In contrast, in the antiferromagnetic and ferromagnetic gapped phases S is finite and scales with β.

Finally, in Fig. 7, we show the temperature dependence of the thermal conductivity κ as a function of magnetic field and separately the contribution from the thermal current D_{QQ} and the magnetothermal contribution MTC (an extensive discussion of the thermal Drude weight as a function of anisotropy and temperature was presented in [32]). Here, in contrast to the spin Seebeck coefficient, κ is strongly suppressed as

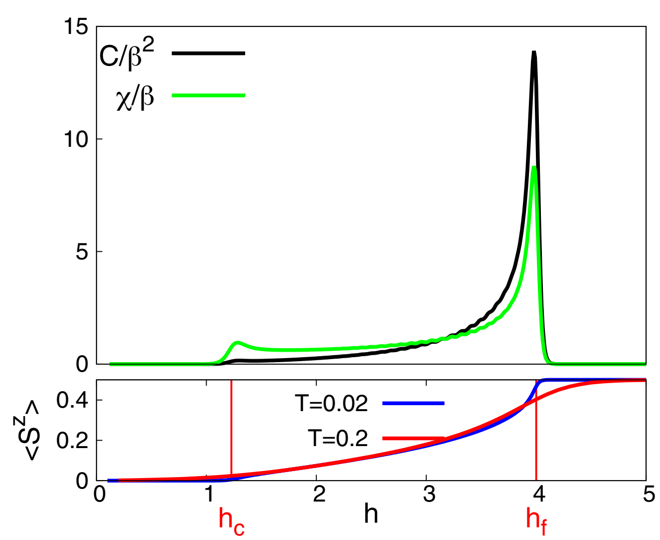

FIG. 5. Specific heat C and magnetic susceptibility χ as a function of magnetic field at temperature $T = 0.02$.

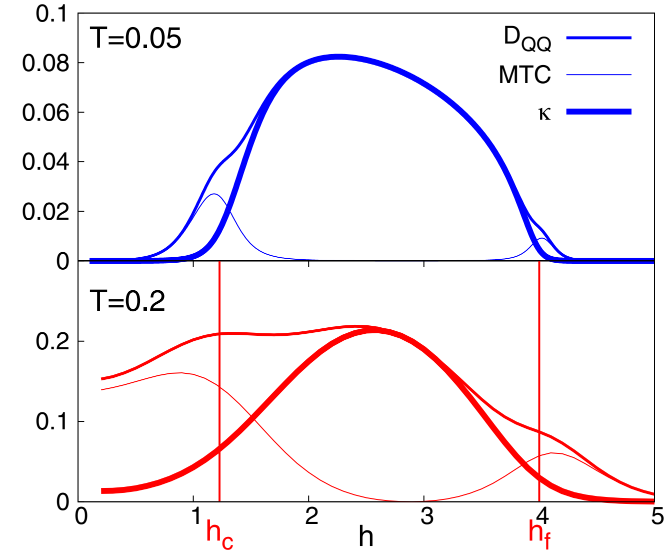

FIG. 7. Thermal conductivity and magnetothermal correction as a function of magnetic field at $T = 0.05$ and $T = 0.2$ for $\Delta = 3$.

expected in the antiferromagnetic and ferromagnetic gapped phases. We also find that the magnetothermal contribution is mostly relevant in the region of the critical fields at low temperatures.

IV. CONCLUSIONS

The main result of this work is that the open boundary (intrinsic, bulk) spin Seebeck effect $\tilde{S}$ in the spin-$\frac{1}{2}$ easy-axis Heisenberg model shows exceptional features at the critical points of the phase diagram in contrast to the usual fixed boundary S coefficient. In other words, the local spin current induced by a local thermal profile has a very different magnetic field dependence from the accumulated magnetization in a fixed boundary system.

Furthermore, the spin-$\frac{1}{2}$ model, although a quantum spin liquid, shares the main features with the classical easy-axis Heisenberg model: in particular, the singular behavior at the critical magnetic fields and the sign change of the spin Seebeck coefficient between the antiferromagnetic and ferromagnetic phase. But it also differs in the diverging spin Seebeck coefficient as $h \to 0$ that we might attribute to the integrability of the model.

Quantum spin liquids are recently becoming candidates for spin current generation in the field of spintronics. $\tilde{S}$ could be studied in experiments aimed at determining phase diagrams and detecting critical points, for instance, in compounds as the spin-$\frac{1}{2}$ easy-axis Heisenberg chains $ACo_2V_2O_8$ (A = Sr, Ba, Pb) [17–19]. The experimental challenge is to study the "open boundary" spin Seebeck effect, e.g., by local magnetothermal imaging [33].

Last but not least, further analytical study of this integrable model should clarify the singular behavior of the induced spin current in the vicinity of the critical fields at low temperatures.

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