Diverging magnetothermal response in the one-dimensional Heisenberg chain

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A current of magnetic moments will flow in the spin-1/2 Heisenberg chain in the presence of an external magnetic field B and a temperature gradient ΔT along the chain. We show that this magnetothermal effect is strictly *infinite* for the integrable Heisenberg model in one dimension. We set up the response formalism and derive several new generalized Einstein relations for this magnetothermal effect which vanishes in the absence of an external magnetic field. We estimate the size of the magnetothermal response by exact diagonalization and quantum Monte Carlo simulations and make contact with recent transport measurements for the one-dimensional Heisenberg compound Sr_2CuO_3.

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I. INTRODUCTION

The nature of magnetic and thermal transport in magnetic insulators with reduced dimensions is a long-standing problem. Huber, in one of the first works on the subject,[1] evaluated the thermal conductivity $\kappa(T)$ for the Heisenberg chain with an equation-of-motion approximation and found a *finite* $\kappa(T)$, a result which is, by now, known to be wrong. A few years later, Niemeijer and van Vianen calculated $\kappa(T)$ in the case $J_z=0$ using the Jordan-Wigner transform and found a diverging result.[2] It has been shown recently[3] that the energy-current operator commutes with the Hamiltonian for the spin-1/2 Heisenberg chain. The thermal conductivity is consequently *infinite* for this model. The intriguing question—"under which circumstances does an interacting quantum system show an infinite thermal conductivity"—is being intensively studied theoretically,[4–6] motivated, in part, by new experimental findings.

An anomalous large magnetic contribution to κ has been observed for the spin-ladder systems $Sr_{14-x}Ca_xCu_{24}O_{41}$ and $Ca_9La_5Cu_{24}O_{41}$,[7,8] raising the possibility of ballistic magnetic transport limited only by residual spin-phonon and impurity scattering. Large energy-relaxation times have been found in recent experimental[9] and theoretical[4] studies of the thermal conductivity for the quasi-one-dimensional spin-chain compounds $SrCuO_2$ and Sr_2CuO_3, consistent with ^{17}O NMR studies.[10]

It is known[3,11] that there is no magnetothermal effect in the Heisenberg chain since the magnetic-current operator is a pseudovector. This can be seen by a simple symmetry argument in the standard setting, when we consider only a temperature gradient along the sample. A nonzero magnetothermal effect would yield a magnetization along an arbitrary quantization axis. But because of the isotropic conditions, there is no preferred direction along which such a magnetization might occur; the effect vanishes consequently.

The situation is, however, different if there is an external magnetic field.[3] A temperature gradient will now cause a magnetization current with a magnetization vector parallel to the field; see Fig. 1. This magnetothermal effect can be considered the generalization to magnetic systems of the Seebeck effect occurring in normal conductors. It is diverging for the isolated Heisenberg chain, leading to a finite nonzero "magnetothermal Drude weight." In this paper we discuss magnetothermal effects in spin chains; i.e., we examine how the magnetic and thermal currents couple to external sources.

In Sec. II we will discuss the differences in between the thermomagnetic effects found in normal metals and the magnetothermal response we study here for magnetic insulators. In Secs. III and IV and in Sec. V we set up the formalism for the magnetic and energy current operators in the presence of an external magnetic field and for the correlation and response functions, respectively.

In Sec. VI we evaluate the magnetothermal response for the xy chain via a Jordan-Wigner transformation and derive and discuss several generalized Einstein relations in Sec. VII. In Sec. VIII we derive and discuss a new exact magnetothermal Einstein relation.

We present in Sec. IX numerical results obtained from quantum Monte Carlo (QMC) simulations and exact-diagonalization studies. Finally, we discuss the size of the magnetothermal effect expected for Sr_2CuO_3 together with a dimensional analysis in Sec. X.

II. THERMOMAGNETIC AND MAGNETOTHERMAL RESPONSE

In a conventional solid there is normally a variety of ways in which external parameters (e.g., current density j, temperature gradient, and magnetic field B) couple to the voltage $\nabla\mu$ and the thermal current j_E leading to numerous thermoelectric and thermomagnetic effects. A phenomenological equation which gives full account of all these effects would look as (see, e.g., Ref. 12)

$$\begin{pmatrix} -\nabla\mu \\ \mathbf{j}_E \end{pmatrix} = \left[\begin{pmatrix} \rho & Q \\ \Pi & -\kappa \end{pmatrix} + \begin{pmatrix} R & N \\ E\kappa & L\kappa \end{pmatrix} \mathbf{B}\times \right] \begin{pmatrix} \mathbf{j} \\ \nabla T \end{pmatrix}.$$

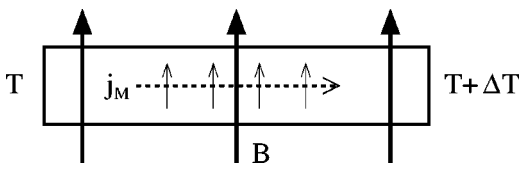

FIG. 1. Illustration of a quasi-one-dimensional magnetic insulator in the presence of an external magnetic field B and a longitudinal temperature differential ΔT. A magnetic current j_M, with the moments aligned along B, is induced.

(**B**$\times$ is understood to be applied to both components separately.) ρ is the resistivity, κ the thermal conductivity, and the other coefficients describe the Seebeck (Q), Peltier (Π), Hall (R), Nernst (N), Ettingshausen (E), and Righi-Leduc effect (L). The latter four effects are called thermomagnetic—for obvious reasons—and are not to be confounded with the effects which are to be discussed in this paper which we denote *magnetothermal effects*. One should note that in magnetic systems the magnetic field corresponds to the chemical potential, leaving no analogs for the entries of the second matrix.

Following Ref. 12, we write the dependence of the current densities j_M (particle) and j_E (energy) on external sources (field $\nabla B = -\nabla\mu$ and temperature gradient ∇T) in the following manner:

$$\begin{pmatrix} j_M \\ j_E \end{pmatrix} = \begin{pmatrix} \hat{L}_{MM} & \hat{L}_{ME} \\ \hat{L}_{EM} & \hat{L}_{EE} \end{pmatrix} \begin{pmatrix} \nabla B \\ (-\nabla T) \end{pmatrix}. \tag{1}$$

[We deviate from the standard notation by indexing the components of the tensor by M and E rather than 1 and 2. The notation with M as particle index and B as chemical potential borrows from spin systems, this article's chief case of interest. Equation (1) may therefore be used as a reference for both electrical and magnetic systems.] Normally, the two off-diagonal components governing the *Seebeck* and *Peltier* effects are not independent but related via the *Onsager* relation. Here, with the given choice of external forces, it reads (where T denotes temperature)

$$T\hat{L}_{ME} = \hat{L}_{EM}. \tag{2}$$

To compute the response functions $\hat{L}_{ij}$ we use the standard Kubo formula[2,6,14,13] which yields for the Heisenberg model generally an $\hat{L}$ tensor with infinite components, i.e., a δ function times a weight factor. These weight factors will be denoted by the entries of the L tensor L_{ij} (without the caret). Normally, one has an additional finite contribution—the regular part—which will turn out to be zero for all but the MM component. We assume that due to some—so far unaccounted for—scattering processes with external degrees of freedoms (like phonons or impurities), the infinite peak broadens and the coefficients (we consider only the real part) may be replaced according to (cf., e.g., Ref. 14)

$$\hat{L}_{ij}(\omega) \equiv L_{ij}\pi\delta(\omega) \to \frac{L_{ij}\tau}{1+(\omega\tau)^2}, \tag{3}$$

with $i,j \in \{M,E\}$ and a finite relaxation time τ—for simplicity, a possible dependence on i and j shall be neglected.

III. CURRENT OPERATORS

In this article, we discuss the analog of the thermoelectric effect in spin chains. The Hamiltonian is the standard xxz chain $H = \Sigma_n h_n$ with

$$h_n = J_x(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y) + J_z S_n^z S_{n+1}^z + \mu_B B S_n^z. \tag{4}$$

Here μ_B is the Bohr magneton and the lattice spacing will be denoted by c; the S_n are supposed to be dimensionless.

Instead of the electrical current we consider the magnetic current[15]

$$J_M = i\mu_B \frac{c}{\hbar}\frac{J_x}{2}\sum_n [S_n^+ S_{n+1}^- - S_n^- S_{n+1}^+]$$

$$= J_x \mu_B \frac{c}{\hbar}\sum_n [\mathbf{S}_n \times \mathbf{S}_{n+1}]_z \equiv i\frac{c}{\hbar}[H, P_M], \tag{5}$$

where we have defined the magnetic polarization $P_M = \mu_B \Sigma_n n S_n^z$, which is possible only for chains with open boundary conditions (OBC's).

For the energy current[2,3] we have equivalently

$$\frac{\hbar}{c}J_E = i\sum_n [h_n, h_{n+1}] = B\frac{\hbar}{c}J_M + \sum_n \tilde{\mathbf{S}}_n \cdot (\mathbf{S}_{n+1} \times \tilde{\mathbf{S}}_{n+2})$$

$$= B\frac{\hbar}{c}J_M + \sum_n \det[\tilde{\mathbf{S}}_n \mathbf{S}_{n+1} \tilde{\mathbf{S}}_{n+2}] \equiv i[H, P_E], \tag{6}$$

where we have used $\mathbf{S}_n = (S_n^x, S_n^y, S_n^z)$ and the definition $\tilde{\mathbf{S}}_n = (J_x S_n^x, J_y S_n^y, J_z S_n^z)$. In the following we will denote with $j_i \equiv J_i/\mathrm{Vol}$, $i \in \{M,E\}$, the respective current densities. Here $\mathrm{Vol} = cN$ is the one-dimensional volume, where c is the lattice constant and N the number of sites.

The energy polarization $P_E = \Sigma_n n h_n$ entering Eq. (6) for OBC's represents a temperature gradient and, as is well known,[16] entails always a magnetic polarization P_M, which stems from the chemical potential term (magnetic field term). To see this we insert a site-dependent (linear)

$$\beta = \beta(n) = \overline{\beta} + nc\nabla\beta$$

into the thermodynamical expectation value.[13] The Boltzmann factor can be rewritten as follows:

$$\exp\left(-\sum_n \beta(n) h_n\right) = \exp\left[-\overline{\beta}\left(H + \frac{c\nabla\beta}{\overline{\beta}}\sum_n n h_n\right)\right]$$

$$= \exp[-\overline{\beta}(H + FcP_E)], \tag{7}$$

where $F = \nabla\beta/\overline{\beta} = -(\nabla T)/\overline{T}$. This motivates the use of P_E to model a temperature gradient.

IV. CORRELATION FUNCTIONS

Setting up the notation for the response theory we define with[17,13]

$$\Lambda(AB)(z) \equiv \frac{i}{\hbar}\int_0^\infty e^{izt}\langle [A(t), B]\rangle dt \tag{8}$$

the retarded Green's function of two operators A and B. In our case A and B will be mostly current operators; we therefore introduce the notation $\Lambda(J_i J_j) \equiv \Lambda_{ij}$. Here $\Lambda(AB)(z)$ vanishes when one of the operators is a constant of motion and commutes with the Boltzmann factor: by the cyclic property of the trace one has in this case $\langle AB\rangle = \langle BA\rangle$.

The energy-current operator J_E commutes with the Hamiltonian in the absence of an external magnetic field B. Using Eq. (6) we have $\Lambda_{ME} = \Lambda_{EM} = B\Lambda_{MM}$ and $\Lambda_{EE} = B^2\Lambda_{MM}$.

The isothermal susceptibility[13] is given by

$$\chi^T(AB) \equiv \int_0^\beta \langle \Delta A(\tau) \Delta B \rangle d\tau, \tag{9}$$

with $\Delta A = A - \langle A \rangle$ and $\Delta B = B - \langle B \rangle$. It takes the usual form $\chi^T(AB) = \beta \langle \Delta A \Delta B \rangle$ when one of the operators is a constant of motion. The generalized Drude weight (which is used, e.g., in Ref. 4) is defined by

$$\langle\langle AB \rangle\rangle \equiv \lim_{z \to 0} (-iz) \int_0^\infty e^{itz} \langle \Delta A(t) \Delta B \rangle dt. \tag{10}$$

These three correlation functions are not independent. Comparing the respective eigenstates-representation, e.g., $Z\Lambda(AB)(0) = \Sigma_{E_n \neq E_k} \Delta A_{nk} \Delta B_{kn} (e^{-\beta E_k} - e^{-\beta E_n})/(E_n - E_k)$, where Z is the partition function, one finds

$$\beta \langle\langle AB \rangle\rangle + \Lambda(AB)(0) = \chi^T(AB). \tag{11}$$

Under open boundary conditions, when the relations $J_j = i(c/\hbar)[H, P_j]$ are valid, we can express the isothermal susceptibility, Eq. (9),

$$\chi^{T,OBC}(J_i J_j) = i \frac{c}{\hbar} \langle [J_i, P_j] \rangle \tag{12}$$

as a static expectation value. Note, however, that the value of the susceptibility is independent of the boundary conditions in the thermodynamic limit: $\chi^{T,OBC}(J_i J_j) = \chi^{T,PBC}(J_i J_j)$.

V. MAGNETOTHERMAL COEFFICIENTS

We now discuss the general recipe for the computation of the entries of the L tensor, appearing in Eq. (3).

We assume a perturbation in the form of a polarization (cf., e.g., Ref. 13), i.e., we add to the Hamiltonian a term $c \nabla B \cdot P_M$ with $P_M := \mu_B \Sigma_n n S_n^z$ and compute the response of the current density operator to obtain the MM component. (A different but equivalent approach is given in Ref. 17.) To compute the remaining entries of the L tensor, we replace the magnetic by the energy current density (in the second row) and/or substitute $P_E := \Sigma_n n h_n$ for P_M—as well as $c \nabla T$ for $c \nabla B$—and add a factor $k_B \beta$ (in the second column). [One should compare this procedure with Eq. (7).]

The linear response theory gives the following contribution to $\hat{L}_{ij}$ (Ref. 13):

$$(k_B \beta)^{j-1} \frac{c}{\hbar} \int_0^\infty e^{izt} \langle i[j_i, P_j(-t)] \rangle dt,$$

where $\beta^M := \beta$, $\beta^E := \beta^2$, etc., and $j_i = J_i/\mathrm{Vol}$ (see Sec. III). Integration by parts yields for the above expression

$$\lim_{z \to 0} \frac{(k_B \beta)^{j-1}}{-iz} \left[i \frac{c}{\hbar} \langle [j_i, P_j] \rangle - \underbrace{\frac{i}{\hbar} \int_0^\infty e^{izt} \langle [j_i(t), J_j] \rangle dt}_{\Lambda_{ij}(z)} \right].$$

One should note that $\Lambda_{ij}(0) = 0$ if one of the currents commutes with the Hamiltonian, and consequently there is no regular part. The factor $(-iz)^{-1}$ accounts for the δ function in the zero-frequency limit and we have

$$L_{ij} = (k_B \beta)^{j-1} \left\{ i \frac{c}{\hbar} \langle [j_i, P_j] \rangle - \Lambda_{ij}(0) \right\}. \tag{13}$$

It is also possible to express this result by the correlation function defined in Eq. (10) using Eqs. (11) and (12):

$$L_{ij} = (k_B \beta)^{j-1} [\chi^T(j_i J_j) - \Lambda_{ij}(0)] = k_B^{j-1} \beta^j \langle\langle j_i J_j \rangle\rangle. \tag{14}$$

From Eq. (13) follows the well-known result for the magnetic Drude weight $L_{MM} = \langle -T_{MM} \rangle c/\hbar - \Lambda_{MM}$, where $T_{MM} = -i[j_M, P_M]$ is the kinetic energy per site—apart from a prefactor. In view of Eq. (14) we define the *generalized kinetic energy*

$$T_{ij} = -i[j_i(B=0), P_j(B=0)]. \tag{15}$$

Here one should note a particularity produced by the twofold role played by the operator P_E in the xxz chain: it not only describes thermal response but also acts as a ''boost'' operator for the constants of motion.[18] Hence, J_E and T_{EE} are constants of motion.

Taking into account that J_E commutes with the Hamiltonian for $B=0$ and using Eqs. (6) and (13) we have

$$\begin{aligned} L_{EM} &= i \frac{c}{\hbar} \langle [j_E, P_M] \rangle - B \Lambda_{MM} = i \frac{c}{\hbar} \langle [j_E(B=0), P_M] \rangle \\ &\quad + B \left\{ i \frac{c}{\hbar} \langle [j_M, P_M] \rangle - \Lambda_{MM} \right\} \equiv \langle -T_{EM} \rangle \frac{c}{\hbar} + B L_{MM} \end{aligned} \tag{16}$$

and at finite B: $\hbar L_{EE}/(c k_B \beta) = \{ i \langle [j_E(B=0) + B j_M, P_E(B=0) + B P_M] \rangle - B^2 \Lambda_{MM} \} = B \langle -T_{ME} \rangle + B \langle -T_{EM} \rangle - \langle T_{EE} \rangle - B^2 \{ \langle T_{MM} \rangle + \hbar \Lambda_{MM}/c \}$.

The generalized kinetic energy $T_{EM} = -i[j_E, P_M]$ appearing in Eq. (16) is given by (in units $\mu_B c/\hbar$)

$$\begin{aligned} T_{EM} &= \frac{-i}{\mathrm{Vol}} \sum_n (n \epsilon_{\alpha\beta\gamma} [\tilde{S}_n^\alpha, S_n^z] S_{n+1}^\beta \tilde{S}_{n+2}^\gamma + \mathrm{cycl.}) \\ &= \sum_n \{ n \tilde{\mathbf{S}}_n \times (\mathbf{S}_{n+1} \times \tilde{\mathbf{S}}_{n+2}) + \mathrm{cycl.} \}_z / \mathrm{Vol} \\ &= \frac{1}{\mathrm{Vol}} \sum_n \{ \tilde{\mathbf{S}}_{n+2} \times (\tilde{\mathbf{S}}_n \times \mathbf{S}_{n+1}) - \tilde{\mathbf{S}}_n \times (\mathbf{S}_{n+1} \times \tilde{\mathbf{S}}_{n+2}) \}_z \\ &= \frac{J_x^2}{\mathrm{Vol}} \sum_n \left\{ -S_n^+ S_{n+1}^z S_{n+2}^- - S_n^- S_{n+1}^z S_{n+2}^+ \right. \\ &\quad + \frac{J_z}{2J_x} (S_n^+ S_{n+1}^- S_{n+2}^z + S_n^- S_{n+1}^+ S_{n+2}^z \\ &\quad \left. + S_n^z S_{n+1}^- S_{n+2}^+ + S_n^z S_{n+1}^+ S_{n+2}^-) \right\}, \end{aligned} \tag{17}$$

where "cycl." means cyclic permutation of $\{n,n+1,n+2\}$—note that the tilde is permuted with the indices.

$\langle -T_{EM}\rangle$ changes sign under inversion of the S^z magnetization M and vanishes consequently for $B=0$, as does the magnetothermal response in this case; compare Eq. (16). The case of a finite B is therefore of interest. We will consider only the effect linear in B, since magnetic fields amount normally only to small energies:

$$L_{EM}\approx B\frac{\partial}{\partial B}L_{EM}|_{B=0}=B\left[\langle -T_{EM}(-\beta M)\rangle\frac{c}{\hbar}+L_{MM}\right]. \tag{18}$$

The EM coefficient, therefore, consists of two parts. One is the B derivative of the zero-field L_{EM}; the other comes from the change of J_E in magnetic field (namely, it acquires a term BJ) and is already linear in B—for small B.

The quotient of L_{ME} and L_{MM},

$$Q_M=\hat{L}_{ME}/\hat{L}_{MM}=L_{ME}/L_{MM}, \tag{19}$$

is called the *(magnetic) thermopower*. It has the advantage that geometric properties of samples cancel in experimental studies and that the relaxation times cancel.

To discuss L_{ME} is superfluous, if one keeps the Onsager relation in mind. Anyway, if we follow the same steps as above, we arrive—with explicitly using OBC's—at (in units $\mu_B c/\hbar$)

$$\begin{aligned}
T_{ME}&=-i[j_M,P_E]\\
&=\frac{-i}{\mathrm{Vol}}\sum_n\ (n[\tilde{\mathbf{S}}_n\times\mathbf{S}_{n+1},\tilde{\mathbf{S}}_n\cdot\mathbf{S}_{n+1}]+(n+1)\tilde{\mathbf{S}}_n\\
&\quad\times[\mathbf{S}_{n+1},\mathbf{S}_{n+1}\cdot\tilde{\mathbf{S}}_{n+2}]+n[\mathbf{S}_{n+1},\tilde{\mathbf{S}}_n\cdot\mathbf{S}_{n+1}]\times\tilde{\mathbf{S}}_{n+2})_z\\
&=\sum\ \{n/2J_x^2(\mathbf{S}_n-\mathbf{S}_{n+1})-(n+1)\tilde{\mathbf{S}}_n\times(\mathbf{S}_{n+1}\times\tilde{\mathbf{S}}_{n+2})\\
&\quad-n\tilde{\mathbf{S}}_{n+2}\times(\tilde{\mathbf{S}}_n\times\mathbf{S}_{n+1})\}_z/\mathrm{Vol}\\
&=\sum_n\ J_x^2/2\{-S_n^+S_{n+1}^zS_{n+2}^--S_n^-S_{n+1}^zS_{n+2}^+\\
&\quad+(n+1)J_z/J_x(S_n^+S_{n+1}^-S_{n+2}^z+S_n^-S_{n+1}^+S_{n+2}^z)\\
&\quad-nJ_z/J_x(S_n^zS_{n+1}^-S_{n+2}^++S_n^zS_{n+1}^+S_{n+2}^-)\\
&\quad+M/\mu_B-S_0^z\cdot\mathrm{Vol}/c\}/\mathrm{Vol},
\end{aligned}$$

which differs apparently from T_{EM}. This does not contradict the Onsager relation because we find that $\langle T_{ME}\rangle=\langle T_{EM}\rangle$ still holds. This may be seen by first using Eqs. (5) and (6),

$$\langle T_{ME}\rangle-\langle T_{EM}\rangle=\frac{c}{\hbar}\langle[[H,P_M],P_E]-[[H,P_E],P_M]\rangle,$$

and then adding a term which is zero by the cyclic property of the trace:

$$=\frac{c}{\hbar}\langle[[H,P_M],P_E]+[[P_M,P_E],H]+[[P_E,H],P_M]\rangle.$$

That this final expression is zero is just Jacobi's identity.

Here it should be emphasized that $\langle T_{ME}\rangle$ has nonnegligible contributions from the boundary and is—unlike $\langle T_{EM}\rangle$—sensitive to a change from periodic boundary conditions (PBC's) to OBC's.

VI. JORDAN-WIGNER TRANSFORM AND FREE FERMION MODEL

For our spin Hamiltonian all quantities may easily be calculated when $J_z=0$ via the Jordan-Wigner mapping to a free fermion system. Under this condition the Hamiltonian is straightforwardly diagonalized by a Fourier transform with eigenvalues $\cos k$ (setting $J_x\equiv 1$) following the Fermi-Dirac distribution $\langle n_k\rangle=\langle c_k^\dagger c_k\rangle=[1+\exp(\beta\cos k)]^{-1}$.

The Drude weight entering Eq. (18) is given simply by $-L_{MM}=\langle T_{MM}\rangle c/\hbar=\int\cos k\langle n_k\rangle dk/(2\pi c)(\mu_B^2 c/\hbar^2)$. At the same time T_{EM} simplifies to (again in units $\mu_B c/\hbar$)

$$\begin{aligned}
T_{EM}&=\frac{1}{\mathrm{Vol}}\sum_n\ \{-S_n^+S_{n+1}^zS_{n+2}^--S_n^-S_{n+1}^zS_{n+2}^+\}\\
&=-\int\frac{dk}{2\pi c}\cos(2k)n_k.
\end{aligned}$$

Following Eq. (18) we need to compute (right-hand side in units $\mu_B^2 c/\hbar$)

$$\langle(-\beta M)(-T_{EM})\rangle=\frac{-\beta}{\mathrm{Vol}}\sum_{k,q}\ \cos(2k)\left\langle n_k\left(n_q-\frac{1}{2}\right)\right\rangle.$$

Here $\langle n_kn_q\rangle=\langle n_k\rangle\langle n_q\rangle$ holds for $k\neq q$, $n_k^2=n_k$ and $\Sigma_{k,q}\cos(2k)\langle n_k\rangle\langle n_q\rangle=0$. We find

$$\begin{aligned}
\langle(-\beta M)(-T_{EM})\rangle&=\frac{\beta}{\mathrm{Vol}}\sum_k\ [-\cos(2k)][\langle n_k\rangle-\langle n_k\rangle^2]\\
&=-\beta\int\frac{dk}{2\pi c}\frac{\cos(2k)}{4\cosh^2[\beta\cos(k)/2]}.
\end{aligned} \tag{20}$$

This result can be related to the temperature derivative of the kinetic energy,

$$\langle T_{EM}(-\beta M)\rangle=\frac{d}{dT}[T\langle -T_{MM}\rangle], \tag{21}$$

where T_{MM} for the xy model is defined above. The relation (21) is easily verified using a partial integration.

VII. EINSTEIN RELATIONS

Let us recapitulate Einstein's relation for diffusive transport in a metal: In a closed system the *diffusive* current $j_D=-\hat{D}\nabla n$, driven by a gradient in the particle density n and the electrical current driven by an *external potential*, j

$=\hat{\sigma}E$, add to zero: $j_D=-j$. This condition yields Einstein's famous relation

$$\frac{\hat{\sigma}}{\hat{D}}=\frac{\nabla n}{E}. \tag{22}$$

Here in the case of a magnetic system, we are not interested in a variation of the electron particle density, but in a change in internal energy or magnetization. Hence, we replace $n(x)\to A_n$ where A_n is either h_n or $\mu_B S_n^z$. Furthermore, the currents are not driven by a field E, but by a polarization $P_F=\Sigma_n nF_n$ (times the lattice constant c), for a conserved F: $[F,H]=0$. In our case the operator F is either the magnetization or the internal energy.

We will also assume that there is diffusive transport for both the magnetization and the energy current. Thereby, we are led to the following definitions of the corresponding diffusion coefficients:

$$j_M\equiv\hat{D}_{MM}\nabla M,\quad j_E\equiv\hat{D}_{EE}\nabla E,$$

where M and E are the magnetization and internal energy.

Generalized Einstein relations can be derived using the *static response* of $\nabla A|_n:=(A_{n+1}-A_n)/c$ to a perturbation cP_F, where c is the lattice constant along the chain:

$$\nabla n/E\to\sum_n \chi^T(\nabla A|_n cP_F)/\mathrm{Vol},$$

where we perform a volume average. (This becomes mandatory because we consider the response to the current density rather than to the current at a fixed site.) Using the linearity of the isothermal susceptibility we find (by a discrete version of an integration by parts)

$$\begin{aligned}c\chi^T(\nabla A|_n P_F)&=\sum_m \{\chi^T[A_{n+1}(mF_m)]-\chi^T[A_n(mF_m)]\}\\&=\sum_m [(m+1)\chi^T(A_nF_m)-m\chi^T(A_nF_m)]\\&=\sum_m \chi^T(A_nF_m)\end{aligned}$$

and with the volume average on both sides

$$\sum_n c\chi^T(\nabla A|_n P_F)/\mathrm{Vol}=\beta\langle\Delta A\Delta F\rangle/\mathrm{Vol}. \tag{23}$$

We rewrite Eq. (22) for the case of a magnetothermal response $j_M\equiv\hat{L}_{ME}\nabla T=\hat{D}_{MM}\nabla M\equiv -j_D$, and find

$$\begin{aligned}\frac{\hat{L}_{ME}}{\hat{D}_{MM}}=\frac{L_{ME}}{D_{MM}}=\frac{\nabla M}{\nabla T}=\frac{k_B}{k_BT}\frac{\nabla M}{(\nabla T/T)}=k_B\beta\chi^T(\nabla M|_n P_E)\\=\frac{k_B\beta^2}{\mathrm{Vol}}\langle\Delta M\Delta H\rangle,\end{aligned} \tag{24}$$

where we have assumed equal relaxation times τ for $\hat{L}_{ME}=\tau L_{ME}$ and the magnetization diffusion: $\hat{D}_{MM}=\tau D_{MM}$. (This assumption was only made for simplicity. At least, to our knowledge there is no reason why the relaxation times should equal each other. However, the assumption is natural as in both cases the finite relaxation time comes from the fact that magnons lose momentum, and the physical processes responsible for that should be in both cases the same.) Equation (24) leads to

$$\begin{aligned}\frac{L_{ME}}{D_{MM}}=\frac{k_B\beta^2}{\mathrm{Vol}}\langle\Delta M\Delta H\rangle\approx B\frac{d}{dB}\frac{k_B\beta^2}{\mathrm{Vol}}\langle\Delta M\Delta H\rangle\\=\frac{Bk_B\beta^2}{\mathrm{Vol}}\langle\Delta M\Delta M\rangle-\frac{Bk_B\beta^3}{\mathrm{Vol}}\langle\Delta M^2\Delta H\rangle;\end{aligned} \tag{25}$$

for small magnetic fields B, the structure of this equation is similar to Eq. (18). The first term of the right-hand side of Eq. (25) is just $B\chi/T$, where $\chi=\beta\langle\Delta M^2\rangle/\mathrm{Vol}$ is the magnetic susceptibility. The second term of the right-hand side of Eq. (25) simplifies to

$$\frac{Bk_B\beta^3}{\mathrm{Vol}}\frac{d}{d\beta}\langle\Delta M\Delta M\rangle=k_BB\beta^3\frac{d}{d\beta}\frac{\chi}{\beta}=-\frac{B}{T}\frac{d}{dT}[T\chi].$$

We therefore find the new relation

$$\frac{L_{ME}}{D_{MM}}=\frac{B}{T}\chi-\frac{B}{T}\frac{d}{dT}[T\chi]=-B\frac{d\chi}{dT}. \tag{26}$$

Classically, the diffusion constant is $\hat{D}=v^2\tau$, where v is the velocity of the elementary excitations, here the magnon velocity. This leads at low temperatures to a temperature-independent diffusion coefficient $D=\hat{D}/\tau=v^2$ and via Eq. (26) to a vanishing magnetothermal response for $T\to 0$, whenever $\chi(T)$ becomes constant for $T\to 0$. Setting $A=M$ and $F=M$ in the general Einstein relation yields[11,14]

$$L_{MM}/D_{MM}=\chi, \tag{27}$$

and with the choice $A=H=F$ one may obtain[11,6] an analogous relation $L_{EE}/D_{EE}=c_V$.

We will now introduce a formula which we believe to be approximately valid at small T. As stated above, the diffusion constant is known not to vary much near $T=0$. Because of our restriction to low T we assume a temperature-independent diffusion coefficient D_{MM} (see discussion above). Then we can rewrite Eq. (27) as

$$\frac{d}{dT}L_{MM}=\frac{d}{dT}[\chi D_{MM}]\approx D_{MM}\frac{d}{dT}\chi.$$

Inserting this expression into Eq. (26) we obtain

$$(-L_{ME})=B\frac{d}{dT}L_{MM}. \tag{28}$$

In the case of free fermions this is just the result of Eq. (21). In the case of a finite interaction it seems to be correct in the limit $T\to 0$ if we use data from Ref. 21. We provide tests of this relation in Sec. IX.

VIII. EXACT EINSTEIN EQUATION

In Sec. VII we set up several versions of generalized Einstein relations appropriate for magnetothermal response. Those relations may be viewed as a link between the dynamical and static response theory, as they connect corresponding correlation functions by introducing diffusion constants.

Upon closer examination of the right-hand sides of Einstein relations like Eq. (27)—namely, χ, c_V, and $d\chi/dT$—we find that that these are static expectation values of products of ΔM and ΔH operators which could be generated by derivatives. Hence, it is an easy task to establish functional relations between the static correlations, e.g.,

$$T^2\frac{d^2}{dB^2}c_v=\frac{d}{dT}\left[T^2\frac{d}{dT}(T\chi)\right].$$

An intriguing question is whether the analogous equations obtained by switching between static and dynamical responses—$\chi\leftrightarrow L_{MM}$, etc.—could also be valid. For one of these relations, Eq. (28), the range of validity is confined to the low-temperature regime unless $J_z=0$. (A detailed discussion follows in Sec. IX.) One reason for the failure could be the fact that in this particular case one of the correlation functions does not reduce to a thermodynamical expectation value. We therefore focus on a relation between the thermal and magnetothermal response coefficients—both mere thermal expectation values. We claim that the relation

$$T^2\frac{d^2}{dB^2}\langle -T_{EE}\rangle=\frac{d}{dT}[T^2\langle T_{EM}(-\beta M)\rangle] \tag{29}$$

is exact. The proof is provided in the Appendix. This equation could be useful to derive an analytical solution for the magnetothermal response.

The arguments of the proof rely mainly on the fact that one of the currents is a constant of motion. It is by the same line of arguments possible to show that Eq. (28) is exact in the case where $[H,J_M]=0$. Unfortunately, this is a very restricting constraint; only the xy model and the Haldane-Shastry Hamiltonian[19] meet the requirement.

IX. NUMERICAL RESULTS

In the free fermion case L_{EM} is easily accessible by a simple evaluation of the analytic expression (20) and a corresponding one for $L_{MM}=-\langle T_{MM}\rangle$, using Eq. (18). If $J_z\neq 0$, we have to compute a mere thermal expectation value in order to obtain T_{EM}, a task which is tractable to Monte Carlo simulations. Here, we use the stochastic series expansion (SSE), which is a Taylor expansion based Monte Carlo method.[20]

We assume a Hamiltonian of the form $H=\Sigma_n J_n h_n$; we may write the partition function $Z=\Sigma[(-\beta)^m/m!]\times\langle\alpha|\Pi_{i=1}^m J_{\phi_m(i)}h_{\phi_m(i)}|\alpha\rangle$ where the sum runs over all numbers m, all functions $\phi_m:\{1,\ldots,m\}\rightarrow\mathbb{N}$, and S^z eigenbasis states α. The SSE program now samples the products of operators appearing in this expression of Z with their relative weight factors.

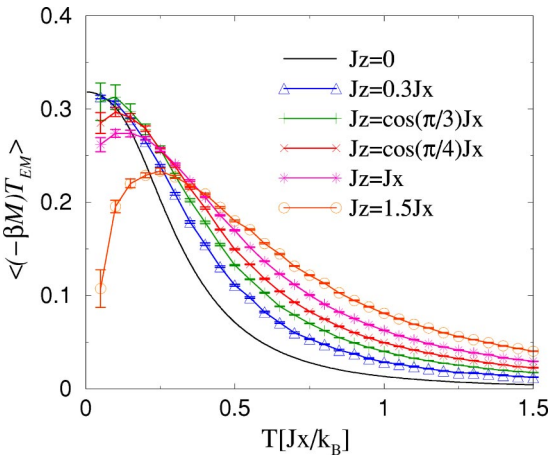

FIG. 2. Monte Carlo data (48 and 96 sites, PBC's) for the dimensionless $\langle(-\beta M)T_{EM}\rangle$ and various interaction strengths J_z/J_{xx}. The statistical MC error bars are given.

Normally, with MC methods it is problematic to measure operators which are not diagonal in the S^z eigenbasis. But with SSE another class of operators is easily accessible: Following a standard procedure in statistical mechanics, we have $\langle h_n\rangle=(\partial_{J_n}Z)/Z$. For the SSE this means that we can measure any operator h_n appearing in the Hamiltonian simply by counting how often it appears in the products sampled by our SSE program (and dividing by J_n). So it is fairly easy to measure sums of products of parts of the Hamiltonian as $S_n^+S_{n+1}^-$, $S_n^-S_{n+1}^+$, or $S_n^zS_{n+1}^z$. To make use of this fact in the given context, it is expedient to find an expression for T_{EM} which consists only of terms appearing in the Hamiltonian. Here we present one (T_{EM} in units $\mu_B c/\hbar$):

$$\begin{aligned}T_{EM}&=\frac{-i\hbar}{\mu_B c}\sum_n\{[j_n,h_{n+1}]+[h_n,j_{n+1}]\}/\mathrm{Vol}\\&=\sum_n\frac{1}{2}\{J_x^2[S_n^+S_{n+1}^--S_n^-S_{n+1}^+,S_{n+1}^+S_{n+2}^-]\\&\quad+J_x^2[S_n^+S_{n+1}^--S_n^-S_{n+1}^+,S_{n+1}^-S_{n+2}^+]+J_xJ_z[S_n^+S_{n+1}^-\\&\quad-S_n^-S_{n+1}^+,S_{n+1}^zS_{n+2}^z]+J_xJ_z[S_n^zS_{n+1}^z,S_{n+1}^+S_{n+2}^-\\&\quad-S_{n+1}^-S_{n+2}^+]\}/\mathrm{Vol},\end{aligned}$$

which then allows us to assess the magnetothermal coefficient by the SSE.

The Drude weight—which is the other input in Eq. (18)—may in principle be calculated by the Bethe-ansatz method. This was attempted by several authors,[22,21] but their calculations are still under discussion,[23] and so far there are no reliable results. We therefore use exact diagonalization to obtain data for the Drude peak. This has clearly the disadvantage that we cannot make any statements about the low-T and high-J_z regimes where the convergence with Vol$\rightarrow\infty$ is slow (cf. Ref. 24).

In Figs. 2 and 3 we plot $\langle(-\beta M)T_{EM}\rangle$ in units of $J_x\mu_B^2/\hbar$—a detailed discussion of units follows in the next section; compare Eqs. (31) and (16)—as a function of tem-

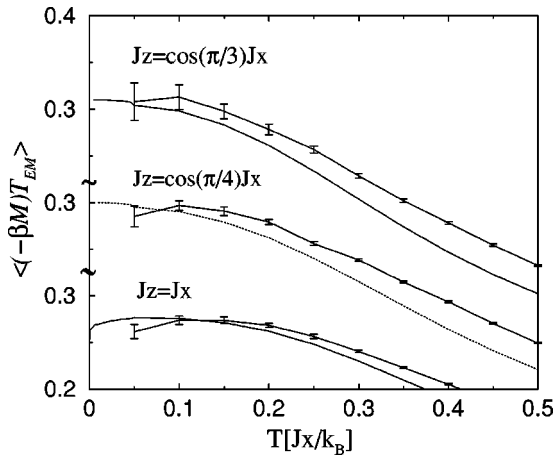

FIG. 3. Monte Carlo data (48 and 96 sites) at small T for PBC's in comparison (lines) with the estimates for $\langle(-\beta M)T_{EM}\rangle$ by Eq. (28) and data from Ref. 21 for the Drude weight. The cases $J_z=\cos(\pi/4)$ and $J_z=1$ are offset for clarity.

perature for various interaction strengths. The data for $J_z\neq 0$ are obtained by MC simulations and—in Fig. 3—by Eqs. (28) and (18).

The MC results in Fig. 2 are clearly not good enough to determine exactly the $T=0$ value. However, for $J_z=J_x$ the data seem to extrapolate to 0.25, which is the Bethe-ansatz result for the dimensionless $T=0$ Drude weight for the isotropic Heisenberg chain.[15] This result would imply via Eq. (18) a vanishing magnetothermal response for $T\rightarrow 0$ and via Eq. (28) a vanishing T derivative of the Drude weight, in contrast to previous results.[22]

In Fig. 3 we present a comparison (at low T) between the MC results for $\langle(-\beta M)T_{EM}\rangle$ and the results from Eq. (28), where we have used Bethe Ansatz results (because of the finite-size gap exact diagonalization provides here no alternative) from Ref. 21—which we prefer to Ref. 22 because it is in agreement with Ref. 23—for the temperature-dependent Drude weight L_{MM}. The agreement is very good for $T<0.2$. As a consequence we may deduce from Eq. (28) that $\langle(-\beta M)T_{EM}\rangle=L_{MM}\hbar/c$ at $T=0$; the linear magnetothermal effect as given by Eq. (18) vanishes consequently for $T\rightarrow 0$.

The results for L_{EM} and the thermopower (the prime denotes a derivative with respect to the magnetic field B) $Q'_M=L'_{ME}/L_{MM}$ are presented in Figs. 4 and 5, respectively. Here we use exact diagonalization, because for the computation of the Drude weight QMC methods normally fail at higher T. (The standard proceeding is to extrapolate from the Matsubara frequencies to $\omega=0$ as attempted in Ref. 14. This procedure becomes soon unstable if T—and hence the spacing of the Matsubara frequencies—grows.) We use exact diagonalization for the computation of $\langle T_{EM}\rangle$ as well. Since $\langle T_{EM}\rangle$ converges much faster with system size than the Drude weight, the finite-size error is determined by the Drude weight, so using QMC simulations for $\langle T_{EM}\rangle$ alone would not give better results. On the contrary, if we used MC data, we would introduce the statistical error. For the Drude peak we use Eq. (14) with $\langle\langle j_i J_j\rangle\rangle=\Sigma_{E_n=E_k}\langle n|j_i|k\rangle$

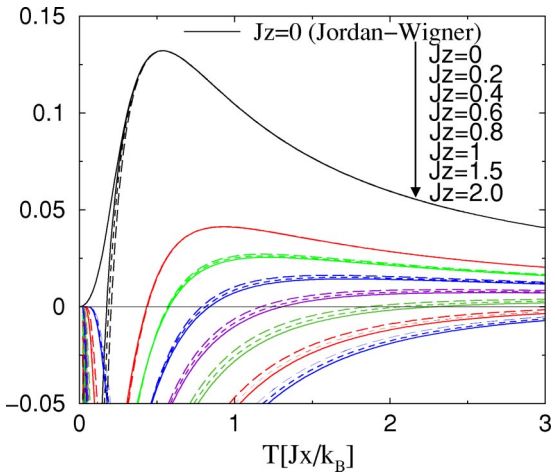

FIG. 4. The linear part (in B) of L_{EM} as a function of temperature [see Eq. (18)] obtained by exact diagonalization for both $\langle(-\beta M)T_{EM}\rangle$ and the Drude weight. We plotted data for three system sizes (14, 16, and 18 sites) with different line styles (long-dashed, dashed, and solid lines, respectively).

$\times\langle k|J_j|n\rangle e^{-\beta E_n}/Z$ (Z is again the partition function); for $\langle T_{EM}\rangle$ we simply use the expression in Eq. (17). The use of these expressions makes it possible for us to exploit translational symmetry which allows us to consider slightly larger systems than in Ref. 24 (namely, up to 18 sites).

Because of the resulting finite-size gap, we can only discuss the high-T regime. (In the xy model we find a vanishing thermopower at $T=0$ with a linear power law.)

For small J_z we find a maximum at intermediate temperatures which decreases and is shifted to higher T as we increase J_z.

At higher J_z the convergence becomes slower, so we cannot make any precise statements about the transport coefficients. However, in Figs. 4 and 5 the curves decrease with system size (for T sufficiently large). This behavior can be confirmed for all systems sizes (smaller than 18 sites). If we

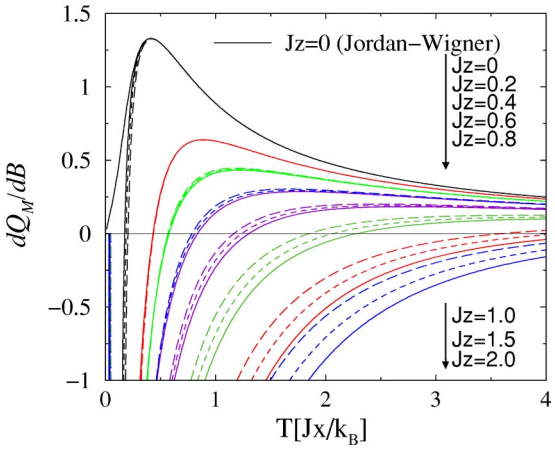

FIG. 5. The part of the thermopower (19) linear in B as a function of temperature [see Eq. (18)] with exact-diagonalization data for $\langle(-\beta M)T_{EM}\rangle$ and the Drude weight. We plotted data for three system sizes (14, 16, and 18 sites) with different line styles (long-dashed, dashed, and solid lines, respectively).

assume that the direction of convergence does not change for larger system sizes, our curves provide an upper bound to the exact results. But this would imply a reversion of the effect (negative thermopower). While at $J_z=0$ the effect is positive—disregarding the low T regime where the negative sign is due to the finite-size gap—one finds $Q'_M \leqslant 0$ for a certain T interval if J_z is large enough. Because of the slow convergence it is impossible for us to predict the precise location of this interval. Here one should emphasize that our conclusions were drawn from the inspection of relatively small systems, so we cannot really exclude that the observed reversion is only due to finite-size effects.

However, we see the reason for this reversion in the fact that the magnetothermal coefficient consists of two summands [see Eq. (18)] which turn out to have opposite signs. These behave very differently under a change of J_z. While the Drude weight dominates at $J_z=0$, it drops dramatically as J_z is increased towards 1 (this was also reported in Ref. 22). The generalized kinetic energy does not decay so much, such that it becomes the dominating part.

X. SIZE OF THE MAGNETOTHERMAL EFFECT

Based on Eqs. (1) and (30) and using the low-B approximation in Eq. (18) we present in this section estimates of the size of the magnetic Seebeck effect.

A. Dimensional analysis

As we are considering effects linear in the magnetic field B we use a prime as a shorthand for $\partial/\partial B$. Using $[J_M]=[\mu_B]$ m/s and $[J_E]=$W m (compare Eqs. (5) and (6) and $[j_{M/E}]=[J_{M/E}]$/m) we may decompose the response coefficients into dimension-full and dimension-less quantities:[4]

$$L'_{ME}=\frac{k_B\mu_B}{\hbar}\frac{\mu_B c}{\hbar}(J_x\beta)^3\tilde{L}'_{ME}, \tag{30}$$

$$L'_{EM}=\frac{J_x\mu_B}{\hbar}\frac{\mu_B c}{\hbar}(J_x\beta)^2\tilde{L}'_{EM}, \tag{31}$$

where the tilde denotes the dimensionless theory result, c is the distance between neighbor sites, and μ_B is the Bohr magneton. Note that L_{ME} has a factor $1/T$ with respect to L_{EM}, hence a third factor $J_x\beta$ in Eq. (30). Inspection of that formula shows the units of L_{ME} to be J T^{-2} m/(K s^2).

For the sake of completeness we state the analogous results

$$L_{MM}=\frac{J_x\mu_B}{\hbar}\frac{\mu_B c}{\hbar}(J_x\beta)\tilde{L}_{MM}$$

for the magnetic Drude weight and

$$L_{EE}=\frac{k_B J_x}{\hbar}\frac{J_x c}{\hbar}(J_x\beta)^2\tilde{L}_{EE}$$

for the thermal Drude weight. The units can be read off as $[L_{MM}]=$J T^{-2} m/s^2 and $[L_{EE}]=$J m/(K s^2). Note that $[\hat{L}_{ij}]=[L_{ij}]$ s; see Eq. (3).

We take the well-studied one-dimensional Heisenberg chain Sr_2CuO_3 as an example,[9] with the following parameters $c=3.91$ Å,[25] $J_x/k_B=2.2\times10^3$ K.[26] There are two chains in an area of 12.68×3.48 Å^2.[25] This leads to a magnon velocity $v_s=J_x c/\hbar\simeq1.2\times10^5$ m/s. The mean free path $\lambda_s(T)$ extracted from a quasiclassical interpretation of thermal conductivity data[9] is strongly temperature dependent; it ranges from $\lambda_s(200\text{ K})\approx100$ Å to $\lambda_s(50\text{ K})\approx800$ Å. This results via $\lambda_s=v_s\tau$ in a relaxation time $\tau(200\text{ K})\simeq0.8\times10^{-13}$ s and $\tau(50\text{ K})\simeq6.6\times10^{-13}$ s, in order of magnitude.

B. Thermopower

For the *thermopower*, we obtain

$$Q'_M=\frac{L'_{ME}}{L_{MM}}=\frac{k_B}{J_x}(J_x\beta)^2\frac{\tilde{L}'_{ME}}{\tilde{L}_{MM}}\equiv\frac{k_B}{J_x}\tilde{Q}'_M.$$

Typically, $k_B/J_x\approx10^{-3}$ K^{-1} (e.g., in Sr_2CuO_3) and at room temperature $\tilde{Q}'_M\approx0.1$. This yields $Q'_M\approx10^{-4}$ K^{-1}.

C. Seebeck effect

The dimensionless B derivative of the magnetothermal response is $(J_x\beta)^3\tilde{L}'_{ME}\simeq0.1$, in order of magnitude; see Fig. 4. We therefore obtain from Eq. (30) at $T=200$ K for the magnetic particle current density a value of

$$0.036\times B\times\nabla T \text{ moments/s}$$

per tesla per (K/m), i.e., in SI units. For a sample of length 1 cm$=10^{-2}$ m with a temperature difference of 10 K and a field of 10 T we have a current of 360 spin-1 moments (magnons) per second. A sample of Sr_2CuO_3 with a cross section of 1 mm^2 contains 4.5×10^{12} chains. It would induce a magnetic current of 1.6×10^{15} moments per second.

D. Closed system

Finally, we consider again the setting of a *closed system*. We are given a sample with open ends such that no current may flow. A temperature gradient should therefore lead to a gradient in the magnetization. Using the Einstein relation for the magnetization current and Eq. (26) we have

$$\nabla M=\frac{L_{ME}}{D_{MM}}\nabla T=-B\frac{\partial\chi}{\partial T}\nabla T.$$

Here $\chi B=M$ is the magnetization caused by the presence of a magnetic field. Hence, dividing by M,

$$\nabla M/M=-\frac{\partial}{\partial T}[\ln\tilde{\chi}]\nabla T,$$

or multiplying both sides with the system length,

$$\Delta M/M=-\frac{k_B}{J_x}\frac{\partial}{\partial\tilde{T}}[\ln\tilde{\chi}]\Delta T.$$

Note that $\Delta M/M$ is not directly a function of magnetothermal coefficients. The magnitude of the dimensionless quantity $(\partial/\partial\tilde{T})[\ln\tilde{\chi}]$ varies about 0.1. We therefore find with $J_x/k_B=2200$ K that the relative change in magnetization—from one end of the sample to the other—should be a fraction of the order of $5\times10^{-5}\mathrm{K}^{-1}\times\Delta T$.

E. Peltier effect

We want to review now briefly the adjoint *(Peltier)* effect, described by the coefficient L_{EM}. We now turn once again to our reference system Sr_2CuO_3. Here it is instructive to compare the energy currents driven by a temperature gradient and a gradient in the magnetic field.

We set all quantities as above, with the exception of the values $(J_x\beta)^2\tilde{L}'_{EM}\simeq0.01$ and $(J_x\beta)^2\tilde{L}'_{EE}\simeq0.1$, which we find more appropriate. We also do not compute the energy current density for one chain but per volume in SI units. The result for the thermal response is

$$j_E=16\times\nabla T\ \mathrm{J/(s\,m^2)}$$

and for the magnetic response

$$j_E=3.3\times10^{-4}\times\nabla B\times B\ \ \mathrm{J/(s\,m^2)}.$$

XI. CONCLUSION

We have shown that nontrivial magnetothermal currents may be induced in one-dimensional quantum-spin chains in an external magnetic field. We have argued that these effects might be especially important for the one-dimensional Heisenberg chain where the energy-current operator commutes with the Hamiltonian and the magnetothermal response diverges. We have presented estimates of the size for the current of magnetic moments induced by a thermal temperature difference for Sr_2CuO_3. We believe the size of the induced magnetic current to be sizeable and note that the observability of magnetic currents in magnetic insulators has been discussed recently.[27]

ACKNOWLEDGMENT

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APPENDIX: PROOF OF Eq. (29)

Proof, first step

We present the proof of Eq. (29) in two steps. First we extend the Hamiltonian to comprise the static response to a thermal current:

$$H(\lambda,B)=H(B)+\lambda J_E.$$

We indicate the expectation value at nonzero λ and B by indices. Moreover, we mean that these variables are zero, if we omit them in an index or an argument. "Δ" means subtraction of the expectation value.

We now show that

$$\langle J_M\Delta H\rangle_{\lambda,B}=\langle J_E\Delta M\rangle_{\lambda,B}. \tag{A1}$$

We explicitly assume PBC's—no polarization operators—and invoke the equation of continuity to obtain a local version of the above formula:

$$\begin{aligned}\langle(J_M^n-J_M^{n-1})\Delta h_m\rangle_{\lambda,B}&=-\langle i[H,\Delta S_n^z]\Delta h_m\rangle_{\lambda,B}\\&=\langle\Delta S_n^z i[H,\Delta h_m]\rangle_{\lambda,B}\\&=-\langle\Delta S_n^z(J_E^m-J_E^{m-1})\rangle_{\lambda,B}.\end{aligned}$$

On the right-hand side of the above equation we perform a reflection in space:

$$\langle(J_M^n-J_M^{n-1})\Delta h_m\rangle_{\lambda,B}=\langle\Delta S_{-n}^z(J_E^{-m}-J_E^{-m+1})\rangle_{-\lambda,B}.$$

Translational invariance leads for

$$a_{n,m}:=\langle J_M^n\Delta h_m\rangle_{\lambda,B},\quad b_{n,m}:=-\langle J_E^n\Delta S_m^z\rangle_{-\lambda,B}$$

to $a_{n,m}\equiv a_{n-m}$ and to

$$a_k-a_{k-1}=b_{k+1}-b_k\Leftrightarrow a_k-b_{k+1}=a_{k-1}-b_k,$$

where $k=n-m$. This last equation implies that

$$c:=a_k-b_{k+1}$$

does not depend on the index k. Our strategy is to show that c—a function on λ and B—may be neglected with impunity in the thermodynamic limit. To this end we look at the following estimate:

$$|c(B,\lambda)|=|b_{k+1}-a_k|\leqslant\min_k\{|b_{k+1}|+|a_k|\}.$$

The fact that all correlation function of the form $\langle\Delta G_1\Delta G_2\rangle$ (such as, e.g., a_k and b_k) decay to zero when the spatial extension (i.e., the index k) goes to infinity leads to $\lim_{\mathrm{Vol}\to\infty}c=0$. Hence we find that in the thermodynamic limit $a_k=b_{k+1}\Rightarrow\Sigma a_k=\Sigma b_k$. And therefore

$$\langle J_MH\rangle_{\lambda,B}/\mathrm{Vol}=-\langle J_EM\rangle_{-\lambda,B}/\mathrm{Vol}.$$

The initial claim (A1) follows—once again—a reflection in space on the second term.

Proof, second step

Having attained our first goal the further proceeding is standard. We apply consecutively derivatives with respect to λ and B on our result and obtain

$$\langle J_MJ_E\Delta H\Delta M\rangle=-T\frac{d}{dB}\langle J_EJ_E\Delta M\rangle_B.$$

We note that the ΔH (ΔM) might come from a derivative with respect to $-d/d\beta=T^2d/dT$ $(-Td/dB)$ and use $\langle T_{EM}(-\beta\Delta M)\rangle=-\beta\langle JJ_E(-\beta\Delta M)\rangle$ and $\langle-T_{EE}\rangle=\beta^2\langle J_EJ_E\rangle$. Our formula may then be rewritten as

$$\frac{d}{dT}[T^2\langle T_{EM}(-\beta M)\rangle]=T^2\frac{d^2}{dB^2}\langle-T_{EE}\rangle_B.$$

Q.E.D.

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