Diffusive Transport in Spin-1 Chains at High Temperatures

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We present a numerical study on the spin and thermal conductivities of the spin-1 Heisenberg chain in the high temperature limit, in particular, the Drude weight contribution and frequency dependence. We use the exact diagonalization and the recently developed microcanonical Lanczos method; it allows us a finite size scaling analysis by the study of significantly larger lattices. This work, pointing to a diffusive rather than ballistic behavior, is discussed with respect to other recent theoretical and experimental studies.

Introduction.—Recently, numerous experiments on quasi-1D spin-1/2 compounds [1–5] have confirmed highly anisotropic thermal transport along the direction of the magnetic chains and a large contribution to the thermal conductivity due to the magnetic interactions. This is in agreement with early theoretical proposals [6,7] of ballistic transport in spin-1/2 Heisenberg antiferromagnetic chains (HAFM), that was recently related to the integrability of this system [8–11]. These developments promoted the theoretical study of several models, such as spin-1/2 frustrated chains, ladders, and higher spin systems, using numerical methods [12–14] or low energy effective theories [15–19].

On the spin-1 compound $AgVP_2S_6$ [20], thermal conductivity experiments revealed anisotropic transport— qualitatively similar to that of spin-1/2 compounds— while NMR [21] concluded to diffusive spin transport at high temperatures and suggested a change in behavior at low temperatures. The $S = 1$ HAFM model is nonintegrable and its physics characterized by a finite excitation gap [22]. Although there has been significant progress in understanding the thermodynamics of $S = 1$ compounds, there are still open questions regarding transport. In particular, theoretical analysis based on a semiclassical approach of the quantum nonlinear sigma model (NLσM) [16,17]—the standard low energy description of $S = 1$ chains and an integrable model—concluded to diffusive dynamics while a Bethe ansatz method calculation [23,24] pointed to ballistic transport.

The present experimental and theoretical status opens two perspectives that motivate this work: First, once the 1D magnetic transport was established as a new mode of thermal conduction, the ongoing synthesis and study of novel compounds demands the theoretical characterization of conductivities—ballistic vs diffusive—in various spin models. Second, the conjectured connection of ballistic (dissipationless) transport to the integrability of

systems requires further theoretical analysis and confirmation.

In this Letter, we present a numerical analysis of the thermal and spin transport properties of the spin-1 HAFM system in an attempt to obtain a first, albeit for finite size lattices, exact picture of the finite temperature/frequency dynamics of this prototype model. We focus the analysis to high temperatures in order to minimize finite size effects and draw reliable conclusions on the thermodynamic limit. In particular, we evaluate the thermal/spin Drude weights, used as the criterion of ballistic or diffusive transport. Additionally, we perform calculations for the spin $\sigma(\omega)$ and thermal $\kappa(\omega)$ conductivity spectra using the exact diagonalization (ED) and the recently developed microcanonical Lanczos method [14,25] (MCLM) which allows us to obtain results for larger systems than hitherto were accessible. The data can be used as a benchmark in the development of analytical theories and in the interpretation of experiments in spin-1 compounds.

Model and method.—The Hamiltonian of the spin-1 HAFM chain is

$$H = J \sum_{l=1}^{L} \mathbf{S}_l \cdot \mathbf{S}_{l+1}, \tag{1}$$

where $\mathbf{S}_l$ is a spin-1 operator at site l and J the exchange constant. We consider periodic boundary conditions and set $J = \hbar = k_B = 1$. The spin j^z and energy j^E current operators obtained from the continuity equations for the local spin S^z and energy H are

$$j^z = J \sum_{l=1}^{L} S_l^x S_{l+1}^y - S_l^y S_{l+1}^x, \tag{2}$$

$$j^E = J^2 \sum_{l=1}^{L} \sum_P (-1)^P S_{l-1}^{P_1} S_l^{P_2} S_{l+1}^{P_3}, \qquad (3)$$

where P are the permutations of x, y, z.

Within linear response theory [26–28] the real part of the thermal conductivity at frequency ω and temperature T is given by

$$\kappa(\omega) = 2\pi D_{\text{th}} \delta(\omega) + \kappa_{\text{reg}}(\omega), \qquad (4)$$

where the regular part of the conductivity κ_{reg} is

$$\kappa_{\text{reg}}(\omega) = \frac{\beta}{\omega L} \tanh\left(\frac{\beta\omega}{2}\right) \Im i \int_0^{+\infty} dt e^{izt} \langle \{j^E(t), j^E\} \rangle, \quad (5)$$

and the thermal Drude weight D_{th} is obtained from

$$D_{\text{th}} = \frac{\beta^2}{2L} \sum_{\substack{n,m \\ \epsilon_n = \epsilon_m}} p_n |\langle m|j^E|n\rangle|^2. \qquad (6)$$

Here $\beta = 1/T$, $z = \omega + i\eta$, p_n are the Boltzmann weights and $|n\rangle$ ($|\epsilon_n\rangle$) the eigenstates (eigenvalues), while in Eq. (5) the symbol $\langle \rangle$ denotes a thermal average. In the $\beta \to 0$ limit we can derive the sum-rule

$$\int_{-\infty}^{+\infty} d\omega \kappa(\omega) = \frac{\pi\beta^2}{L} \langle (j^E)^2 \rangle = I, \qquad (7)$$

suggesting that a measure of the ballistic contribution to the conductivity is given by the quantity $2\pi D_{\text{th}}/I$. The corresponding equations for the regular part of the spin conductivity $\sigma_{\text{reg}}(\omega)$ and Drude weight D can be obtained from Eqs. (5) and (6) above by replacing j^E by j^z and dividing them by β.

Drude weight data are obtained by using ED which restricts us to system sizes up to $L = 12$ sites. We use the translational and spin symmetries of our system to perform the calculation in subspaces of momentum k and magnetization S_{tot}^z. We find that the results obtained in the $k = 0$, $S_{\text{tot}}^z = 0$ subspace for $L = 12$ (space dimension $\simeq 6500$) are very close to those obtained by diagonalizing the entire Hilbert space.

For the high temperature $\kappa(\omega)$ and $\sigma(\omega)$ calculations we employ the MCLM method [14,25] which allows us to obtain results for systems up to $L = 18$ sites. The spectra calculated using this method include the Drude weight as a low frequency peak with width of the order of the frequency resolution of the method; notice however that this contribution is negligible for the larger systems we study as it follows from the finite size scaling of the Drude weights (see Figs. 1 and 4). Here we have used ~ 1000 Lanczos steps for the first Lanczos procedure and ~ 4000 Lanczos steps for the continuous fraction expansion which results in an ω resolution of ~ 0.01.

Thermal conductivity.—In Fig. 1 we show the temperature dependence of the thermal Drude weight for several system sizes L. D_{th} is vanishing at $T = 0$ while at high temperatures it has a simple β^2 dependence. A nonzero
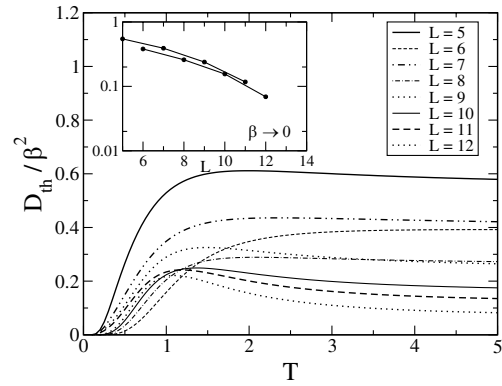

FIG. 1.   The thermal Drude weight as a function of temperature for several system sizes. In the inset, the scaling of D_{th}/β^2 for $\beta \to 0$ for odd and even number chains.

Drude weight is generally expected for systems with size less than the mean free path of the magnetic excitations. In the $\beta \to 0$ limit, as L increases, D_{th}/β^2 decreases—seemingly exponentially fast—and appears to scale to zero in the thermodynamic limit as seen by the curves in the inset of Fig. 1 (for chains with even and odd number of sites). Our data therefore suggest diffusive thermal transport for the spin-1 HAFM chain.

We now apply the MCLM method to calculate the ω dependence of the thermal conductivity $\kappa(\omega)$ in the high temperature limit as shown in Fig. 2. For frequencies $\omega \gtrsim 0.05$ the conductivity obtained for $L = 18$ has practically converged to the $L \to \infty$ limit while in the low frequency regime there is a remaining size dependence. This is partly due to the variation of the Drude weight which contributes to the low frequency $\kappa(\omega)$. It is worth noting that the statistical fluctuations in our MCLM results are very small, even for the smallest size system displayed here. A comparison of ED versus MCLM results for $L = 12$ (not shown for clarity) gives satisfactory agreement although for this size system the statistical fluctuations are significant.

On the low frequency region, it is not well described by a Lorentzian, as predicted by the diffusion phenomenol-
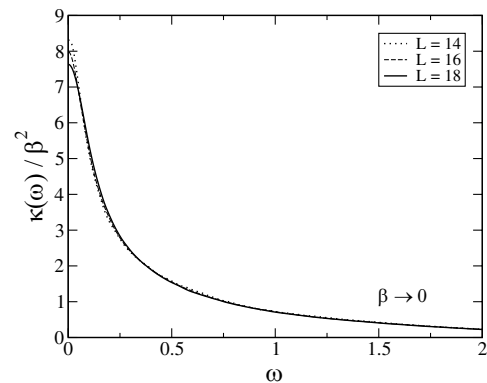

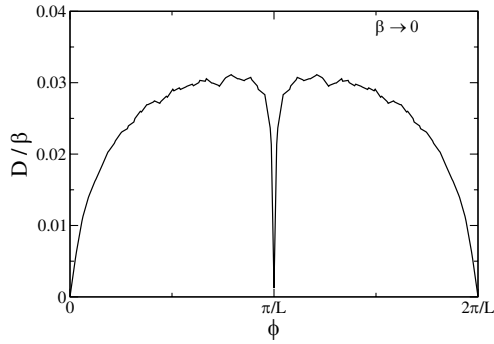FIG. 2.   The thermal conductivity $\kappa(\omega)$ for $\beta \to 0$.

FIG. 3. The ϕ dependence of the Drude weight for $L = 10$.

FIG. 4. Drude weight D as a function of temperature for several system sizes L. In the inset, the scaling of D/β for $\beta \to 0$.

ogy [29], but the overall form of $\kappa(\omega)$ is similar to that found in the $S = 1/2$ ladder model [14] and other low dimensional models [30]; it suggests that this may be a generic behavior of conductivity spectra in such systems. From this curve we can also extract an estimate [14] of the high temperature $\kappa_{dc} = \kappa(\omega \to 0)$ thermal conductivity, $\kappa_{dc} \simeq 16(\beta J)^2 \frac{W}{mk}$, assuming typical lattice constants $O(10 \text{ Å})$ and $J \sim O(1000 \text{ K})$.

For comparison we note that (i) a $\kappa_{dc} \sim O(1\frac{W}{mK})$ was observed at temperatures below the gap, $T \sim 0.2J$, in the compound $AgVP_2S_6$ [20] and (ii) our high temperature κ_{dc} for the spin-1 model is an order of magnitude larger than that of the ladder [14]; notice that the spin-1 HAFM has a similar low energy excitation spectrum and thus low temperature behavior as the spin-1/2 two-leg ladder for $J_\perp/J \simeq 0.9$.

Spin conductivity.—We now investigate the spin transport by calculating the Drude weight D and spin conductivity $\sigma(\omega)$. For $L \leq 12$, D/β appears to be equal to zero (up to numerical precision) at all temperatures. On this issue it is important to point out that, for faster convergence, we consider only the $S^z = 0$ subsector that is the dominant one in the thermodynamic limit. In order to explore the robustness of this result we apply the canonical transformation $S_l^+ \to S_l^+ e^{i\phi l}$ on H and j^z (periodic in ϕ with period $2\pi/L$); the results for D as a function of ϕ, are shown in Fig. 3 for $L = 10$.

We find that D is finite for all ϕ, except for $\phi = 0$ and $\phi = \pi/L$ where it develops a sharp minimum. Curves for different L's show very similar ϕ dependence, but with the local minimum at $\phi = \pi/L$ becoming sharper with increasing L. We therefore conclude that the vanishing Drude weight at $\phi = 0$, even for small L, is an artifact of the periodic boundary conditions; notice that a vanishing D/β and a nontrivial ϕ dependence is also found in the $S = 1/2$ isotropic model.

In Fig. 4 we show that D/β, close to its maximum value at $\phi = \pi/2L$, is finite throughout the temperature range and as shown in the inset, it scales to zero exponentially fast with L in the $\beta \to 0$ limit. In contrast to the thermal Drude weight, D/β goes to a finite value at very low temperatures that can be understood considering that
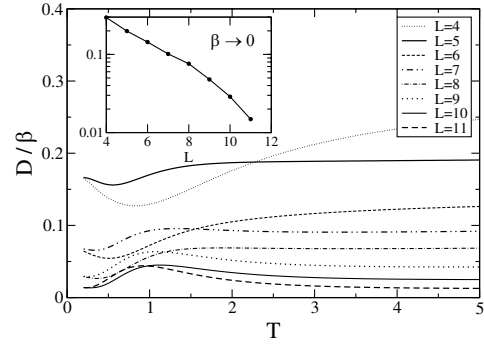
a ϕ implies a ground state carrying a nonzero spin current.

Finally, we present in Fig. 5 $\sigma(\omega)$ evaluated using the MCLM method. We see that there are some statistical fluctuations in the data for the smaller systems while those for the larger systems are very smooth. The curves seem converged to their $L \to \infty$ limit for $\omega \gtrsim 0.2$. The main characteristics of our results is the appearance of a local maximum at $\omega \sim 1$ and a minimum at $\omega \lesssim 0.2$. The later disappears with increasing system size while again we see no signs of a Drude peak in the $\sigma(\omega)$ curves. It is interesting to note that the local maximum feature has also previously been observed in a study of correlations of the NLσM [17].

Discussion.—The overall picture emerging from the presented numerical data shows that the high temperature spin and energy transport of the spin-1 HAFM chain is characterized by finite dc values, vanishing Drude weights, a smooth frequency dependence (though not of a Lorentzian form) and thus nonballistic character. This behavior is compatible with the assumption of normal transport in nonintegrable models: it is qualitatively similar to that of spin-1/2 antiferromagnetic ladder and in contrast to the ballistic transport of the integrable spin-1/2 version. On this point we should mention that in the
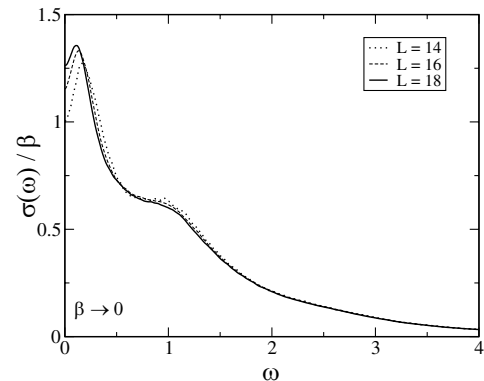

FIG. 5. The spin conductivity $\sigma(\omega)$ for $\beta \to 0$.

isotropic spin-1/2 model D also seems to vanish [8]; however $\sigma(\omega \to 0)$ might diverge [25] and, in any case, in the easy-plane anisotropic $S = 1/2$ model D is finite, (in contrast to preliminary results on the anisotropic $S = 1$ model). On the other hand, for $S = 1/2$, D_{th} is clearly finite, as the energy current operator commutes with the Hamiltonian, again in contrast to the $S = 1$ case.

From our data in Figs. 2 and 5 we can also extract the spin $\mathcal{D}_s = \sigma_{\mathrm{dc}}/\chi \sim 1.4\beta/\frac{2}{3}\beta \sim 2.1$ and thermal $\mathcal{D}_{\mathrm{th}} = \kappa_{\mathrm{dc}}/C \sim 7.5\beta^2/\frac{4}{3}\beta^2 \sim 5.6$ diffusion constant (in units of $J/\hbar$), where χ is the static susceptibility and C the specific heat. In comparison, a standard $\beta \to 0$ moment analysis [6] gives $\mathcal{D}_s = \sqrt{2\pi S(S+1)/3} \sim 2.1$ and $\mathcal{D}_{\mathrm{th}} = \sqrt{\pi S(S+1)/3}/[1 - 3/4S(S+1)] \sim 2.3$; the agreement for $\mathcal{D}_s$ is excellent (also probably fortuitous considering the quantum character of the $S = 1$ system) while an enhanced value is found for $\mathcal{D}_{\mathrm{th}}$.

Regarding the low temperature behavior, the limitation of our calculation to small size systems (thus a sparse low energy spectrum) does not allow us to make any reliable statements and, in particular, to study an eventual change of transport from diffusive to ballistic as suggested by the experimental results for $AgVP_2S_6$ [21]. Yet, there exist known spin-1 compounds, with weak values of J, for which these data are directly relevant in the interpretation of transport experiments. A crucial issue remains however for future studies, namely, the disentanglement of the spin-phonon from the intrinsic spin-spin scattering contribution to diffusion.

Finally, on the low energy NLσM approach [16,23,24], this high temperature study cannot shed light on the issue of ballistic vs diffusive behavior. If it is concluded that the NLσM predicts diffusive transport then there is continuity with the present $\beta \to 0$ data. If, on the other hand, ballistic transport (perhaps due to the integrability of the NLσM) is found, then the omitted "irrelevant" terms (for thermodynamics) could result in a diffusive behavior at all temperature scales.

[1] A. V. Sologubenko, K. Giannó, H. R. Ott, U. Ammerahl, and A. Revcolevschi, Phys. Rev. Lett. **84**, 2714 (2000).
[2] A. V. Sologubenko, K. Giannó, H. R. Ott, A. Vietkine, and A. Revcolevschi, Phys. Rev. B **64**, 054412 (2001).
[3] C. Hess, C. Baumann, U. Ammerahl, B. Büchner, F. Heidrich-Meisner, W. Brenig, and A. Revcolevschi, Phys. Rev. B **64**, 184305 (2001).
[4] K. Kudo, S. Ishikawa, T. Noji, T. Adachi, Y. Koike, K. Maki, S. Tsuji, and K. Kumagai, J. Phys. Soc. Jpn. **70**, 437 (2001).
[5] A. V. Sologubenko, H. R. Ott, and A. Revcolevschi, Europhys. Lett. **62**, 540 (2003).
[6] D. L. Huber and J. S. Semura, Phys. Rev. **182**, 602, (1969); D. L. Huber, J. S. Semura, and C. G. Windsor, Phys. Rev. **186**, 534 (1969).
[7] T. Niemeijer and H. A. W. van Vianen, Phys. Lett. **34A**, 401 (1971).
[8] for a review see cond-mat/0304630 ["Interacting Electrons in Low Dimensions", edited by D. Baeriswyl and L. de Giorgi, (Kluwer, to be published)].
[9] X. Zotos, F. Naef, and P. Prelovšek, Phys. Rev. B **55**, 11029 (1997).
[10] K. Saito, S. Takesue, and S. Miyashita, Phys. Rev. E **54**, 2404 (1996).
[11] B. N. Narozhny, A. Millis, and N. Andrei, Phys. Rev. B **58**, R2921 (1998).
[12] J. V. Alvarez and C. Gros, Phys. Rev. Lett., **89**, 156603 (2002).
[13] F. Heidrich-Meisner, A. Honecker, D. C. Cabra, and W. Brenig, Phys. Rev. B **66**, 140406 (2002); **68**, 134436 (2003).
[14] X. Zotos, Phys. Rev. Lett. **92**, 067202 (2004).
[15] A. Rosch and N. Andrei, Phys. Rev. Lett. **85**, 1092 (2000).
[16] S. Sachdev and K. Damle, Phys. Rev. Lett. **78**, 943 (1997); K. Damle and S. Sachdev, Phys. Rev. B **57**, 8307 (1998).
[17] C. Buragohain and S. Sachdev, Phys. Rev. B **59**, 9285 (1999).
[18] E. Orignac, R. Chitra, and R. Citro, Phys. Rev. B **67**, 134426 (2003).
[19] K. Saito, Phys. Rev. B **67**, 064410 (2003).
[20] A. V. Sologubenko, S. M. Kazakov, and H. H. Ott, Phys. Rev. B **68**, 094432 (2003).
[21] M. Takigawa, T. Asano, Y. Ajiro, M. Mekata, and Y. J. Uemura, Phys. Rev. Lett. **76**, 2173 (1996).
[22] F. D. M. Haldane, Phys. Lett. A **93**, 464 (1983).
[23] S. Fujimoto, J. Phys. Soc. Jpn. **68**, 2810 (1999).
[24] R. M. Konik, Phys. Rev. B **68**, 104435 (2003).
[25] M. W. Long, P. Prelovšek, S. El Shawish, J. Karadamoglou, and X. Zotos, Phys. Rev. B **68**, 235106 (2003).
[26] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[27] J. M. Luttinger, Phys. Rev. **135**, A1505 (1964).
[28] F. Naef and X. Zotos, J. Phys. Condens. Matter **10**, L183 (1998); F. Naef, Ph.D. thesis, EPF-Lausanne [Institution Report No. 2127, 2000 (unpublished)].
[29] L. P. Kadanoff and P. C. Martin, Ann. Phys. (Berlin) **24**, 419 (1963).
[30] J. Jaklič and P. Prelovšek, Adv. Phys. **49**, 1 (2000).