Diffusive energy transport in the $S = 1$ Haldane chain compound AgVP$_2$S$_6$ 

A.V. Sologubenko, S.M. Kazakov and H.R. Ott  
Laboratorium für Festkörperphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland

T. Asano and Y. Ajiro  
Department of Physics, Kyushu University, Fukuoka 812-8581, Japan

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We present the results of measurements of the thermal conductivity $\kappa$ of the spin $S = 1$ chain compound AgVP$_2$S$_6$ in the temperature range between 2 and 300 K and with the heat flow directed either along or perpendicular to the chain direction. The analysis of the anisotropy of the heat transport allowed for the identification of a small but non-negligible magnon contribution $\kappa_m$ along the chains, superimposed on the dominant phonon contribution $\kappa_{ph}$. At temperatures above about 100 K the energy diffusion constant $D_E(T)$, calculated from the $\kappa_m(T)$ data, exhibits similar features as the spin diffusion constant $D_S(T)$, previously measured by NMR. In this regime, the behaviour of both transport parameters is consistent with a diffusion process that is caused by interactions inherent to one-dimensional $S = 1$ spin systems.

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I. INTRODUCTION

Anomalous features of transport properties of one-dimensional Heisenberg antiferromagnetic (1D HAFM) spin $S = 1/2$ systems were predicted decades ago. In particular, it was argued that both the spin and thermal conductivity are not expected to be of diffusive character as is typical for classical systems without long-range order, but instead are based on ballistic transport of spin and energy, at least in the case of ideal 1D systems.\textsuperscript{3,4,5,6} On theoretical grounds it was recently demonstrated that this difference is due to the conservation of spin and energy currents in integrable models which apply for 1D $S=1/2$ spin systems.\textsuperscript{7,8,9,10,11} The situation is much less clear with respect to the theory of transport properties of 1D $S=1$ systems. Contrary to half-integer spin 1D HAFM systems with isotropic exchange, for which the spin excitations are $S = 1/2$ spinons and the corresponding spectrum is gapless,\textsuperscript{12} the integer-spin chains exhibit gapped excitation spectra and $S = 1$ magnon excitations.\textsuperscript{13} Numerical calculations for $S = 1$, 1D HAFM systems relate the gap $\Delta$ to the exchange integral $J$, such that $\Delta = 0.41J$.\textsuperscript{14} Early theoretical work predicted that the transport in ideal 1D HAFM $S=1$ systems must be diffusive.\textsuperscript{3} More recent discussions of Sachdev and Damle,\textsuperscript{15,16} considering the spin transport in Heisenberg $S=1$ chains in terms of the nonlinear $\sigma$-model (NL$\sigma$M), also concluded with the claim of diffusive spin transport. However, Fujimoto,\textsuperscript{17} based on the integrability of NL$\sigma$M, suggested that in a perfect 1D $S=1$ system, the spin transport is ballistic. The author noted, however, that external perturbations, unavoidable in real $S=1$ chain compounds and leading to the destruction of integrability, may restore diffusive transport. In this respect, Fujimoto refers to a previous experimental observation of spin diffusion in the $S=1$ chain compound AgVP$_2$S$_6$, by Takigawa et al.,\textsuperscript{18} and suggests that the spin-phonon interaction might provide this type of perturbation.

Energy transport via spin excitations, which can be probed by measurements of the thermal conductivity, has not yet been investigated in $S=1$ chain compounds. It seems natural to expect that its features are similar to those of spin transport. In a recent paper by Alvarez and Gros,\textsuperscript{19} relations between the spin and thermal conductivities in 1D spin systems, similar to the famous Wiedemann-Franz law for the ratio between the electrical and thermal conductivities in metals, were discussed. In the present work, we report the results of an experimental investigation of the thermal conductivity of AgVP$_2$S$_6$, a compound which is considered as one of the best physical realizations of an $S = 1$ 1D AFM Heisenberg model system. AgVP$_2$S$_6$ crystallizes with a monoclinic crystal structure of type $P2_1/a$.\textsuperscript{20} Each $V^{3+}$ ion with spin $S = 1$ is located inside a distorted octahedron of sulfur ions. The zigzag chains formed by these vanadium ions run along the $a$ axis. Evidence for an energy gap in the spin excitation spectrum was found by measurements of the magnetic susceptibility\textsuperscript{21,22,23,24} and inelastic neutron-scattering experiments.\textsuperscript{25,26} The magnetic properties of the compound are well described by the isotropic nearest-neighbor Heisenberg Hamiltonian and weak single-ion anisotropy

$$H = J \sum_i \mathbf{S}_i \mathbf{S}_{i+1} + D \sum_i (\mathbf{S}_i^z)^2,$$

with the intrachain exchange constant $J/k_B = 780$ K and $D/J = 5.8 \times 10^{-3}$.\textsuperscript{27} The interchain interaction $J' \lesssim 10^{-5}J$ is very weak.\textsuperscript{25} The dynamics of the $V^{3+}$ spins in AgVP$_2$S$_6$ was studied by NMR experiments by Takigawa et al.\textsuperscript{18} The data are compatible with spin diffusion at temperatures exceeding $\sim$ 100 K and above a temperature-dependent frequency of the order of $10^{11} - 10^{12}$ s$^{-1}$. Using these results, the temperature dependence of the spin diffusion constant $D_S(T)$ was established. Below we compare the experimental results...
for the temperature variation of the spin diffusion constant $D_S(T)$, probed by NMR, and the energy diffusion parameter $D_E(T)$, which is obtained from the results of our thermal conductivity measurements.

II. SAMPLES AND EXPERIMENT

For the measurements of the thermal conductivity $\kappa(T)$, we grew several single crystals of AgVP$_2$S$_6$ by solid state reaction of stoichiometric amounts of Ag, V, P, and S, as described in Ref. 20. The largest crystals had dimensions of several mm along the $a$-axis (the spin chain direction), a maximum of 0.5 mm along the $b$-axis and, at most, 0.2 mm along the $c$-axis. A single crystalline specimen with dimensions of $2 \times 0.5 \times 0.15$ mm$^3$ (denoted as S1) was used for the thermal conductivity measurements along the chain direction. Another sample from the same batch served to measure the dc magnetic susceptibility $\chi$.

A standard method for separating the spin-mediated thermal conductivity $\kappa_m(T)$ from other, less anisotropic, contributions to $\kappa(T)$ is based on measurements of this quantity both along and perpendicular to the chain direction. Unfortunately, the dimensions of our crystals only allowed for measurements of $\kappa(T)$ along the chains. For this reason we investigated the heat transport also on grain-aligned specimens. Some of these samples were previously used for measurements of the magnetic susceptibility and NMR experiments. Each of the specimens with typical sizes of $4 \times 2 \times 0.2$ mm$^3$ were single pieces consisting of many equally oriented flake-like single crystals. On one such piece (denoted as sample J1), the thermal conductivity was measured along the $a$-axis. Another sample, denoted as J2, with the largest spatial extension along the $b$-axis, was cut out from the central part of sample J1. Sample J2 was subsequently used for experiments with the heat flow perpendicular to the chain direction.

The thermal conductivity was measured using a standard uniaxial heat flow method, where the constant heat flux along a rectangular-bar shaped sample was produced by a heater attached to one end of the sample. The opposite end of the sample was attached to a copper heat sink. The temperature gradient was monitored by a system of 25 $\mu$m diameter Chromel-Au+0.07%Fe thermocouples. We estimate the uncertainty of the absolute value of $\kappa$ to be of the order of 10%, because of the uncertainty of the sample geometry. The relative error of the measured thermal conductivity, however, is only about 0.5%. The magnetic susceptibility was measured between 4.5 and 240 K, employing a commercial SQUID magnetometer.

III. RESULTS AND DISCUSSION

The magnetic susceptibility data are shown in Fig. 1. The measurements were taken in a magnetic field of $H = 40$ kOe oriented along the $a$-axis. The data are in good agreement with results of Mutka et al., also shown in Fig. 1. In particular, the nonlinear $\chi$ vs. $1/T$ dependence at low temperatures, noticed in Ref. 28 and attributed to the weak coupling between finite-length segments of spin chains, is well reproduced in our data (see the inset of Fig. 1). The $\chi(T)$ data can be fit to the equation

$$\chi(T) = \frac{A}{T^{1/2}} \exp \left( -\frac{\Delta}{k_BT} \right) + \frac{B}{T^\alpha} + \chi_0,$$

where the first term on the right-hand side represents the contribution from the Haldane-gapped $S = 1$ chains, the second term reflects the above mentioned finite segment interaction contribution and the last and constant term is due to the diamagnetic orbital susceptibility. The best fit, shown as the solid line in Fig. 1, is achieved with the fit parameters $A = 1.77 \times 10^{-2}$ emu mol$^{-1}$ K$^{1/2}$, $\Delta/k_B = 244$ K, $B = 3.60 \times 10^{-3}$ emu mol$^{-1}$ K$^\alpha$, $\alpha = 0.55$, and $\chi_0 = -2.5 \times 10^{-4}$ emu mol$^{-1}$. The resulting value for the energy gap $\Delta/k_B$ is consistent with estimates of 228 K (Ref. 22) and 250 K (Ref. 21) from the analyses of magnetic susceptibilities, but somewhat lower than the results obtained from NMR (320 K) and the neutron scattering measurements. The power-law exponent $\alpha = 0.55$ is within the range 0.5 < $\alpha$ < 0.8 established in Ref. 22.

The temperature dependences of the different thermal conductivities are shown in Fig. 2. Each curve exhibits a maximum at temperatures around 10 K and a region with a positive slope $d\kappa/dT$ at temperatures above about 200 K. The latter feature is presumably due to unaccounted heat losses via radiation, typical in standard steady-state thermal conductivity experiments probing materials with low values of $\kappa$ at high temperatures. Our
numerical estimates suggest that for our AgVP$_2$S$_6$ samples, these effects are negligible only below about 170 to 200 K and therefore, our data analysis presented below is restricted to temperatures below 170 K.

By rapidly reducing the temperature of the grain-aligned samples (J1 and J2) from values in the region between 170 and 300 K to temperatures between 80 and 170 K with a cooling rate of approximately 1 K/s, subsequent slow relaxations of $\kappa$ with time were observed. As illustrated in the inset of Fig. 2, even upon a slow variation of the temperature (as slow as 0.01 K/s), a small hysteresis of $\kappa(T)$ was observed at intermediate temperatures above the maximum of $\kappa(T)$. We believe that these effects are not related to intrinsic properties of the material. Since they were not observed when probing the single-crystalline specimen, they are most likely due to the multigrain nature of these samples.

By inspecting the $\kappa(T)$ curves at temperatures below 60 K, we note that they all exhibit the same qualitative features. At higher temperatures, a shoulder-type feature in $\kappa_a(T)$, which is absent in the $\kappa_b(T)$ curve, may be identified. Similar high-temperature features emerging as a shoulder or a second maximum were observed for $\kappa(T)$ if measured along the chain or ladder direction of spin-chain and spin-ladder compounds.$^{29,30,31,32,33,34,35,36}$ An obvious interpretation of this high-temperature feature is to ascribe it to a spin-mediated thermal conductivity $\kappa_m$, in addition to the common heat conduction via phonons, which is reflected in a contribution $\kappa_{ph}$ to the total thermal conductivity.

In order to isolate the spin contribution $\kappa_m(T)$, accurate evaluations of other contributions to the total measured $\kappa(T)$ need to be made. Since AgVP$_2$S$_6$ is an insulator, no heat conduction by free charge carriers is expected, and only heat transport via lattice excitations needs to be considered. Because of the extremely weak interaction between spins of neighboring chains, no sizable heat transport via the spin system is expected for directions perpendicular to the $a$-axis. Hence only phonons contribute to $\kappa_b(T)$, shown in Fig. 2. The shape of the $\kappa_b(T)$ curve, typical for phonon heat transport, results from the competition between different phonon scattering mechanisms, most notably phonon scattering by sample or grain boundaries at the lowest temperatures and by phonons, defects and magnons at more elevated temperatures. In contrast to the boundary and defect scattering which is usually sample-dependent, the scattering of phonons by quasiparticles in general is expected to be sample-independent because it is related to intrinsic scattering processes.

At temperatures much less than $\Delta/k_B$, the number of spin excitations that participate in the heat transport reflected in $\kappa_a(T)$ is decreasing exponentially with decreasing temperature. Therefore, at $T \ll \Delta/k_B$ the phonon heat transport is expected to also dominate $\kappa_a(T)$. While the different temperature dependences of $\kappa$ for different samples on the left of the $\kappa(T)$ maxima in Fig. 2 can be accounted for by differences in the boundary scattering conditions, it is remarkable that at temperatures above the maxima, up to about 40 K, the ratio between the $\kappa$ values for different samples is practically independent of temperature. This implies that the temperature dependence of these $\kappa(T)$ curves are identical and that in this temperature region, the intrinsic processes of phonon scattering dominate. As we have already argued in Ref. 29, the anisotropy of the purely phononic heat transport is not likely to change at high temperatures. This is why we assume that at high temperatures, the phonon thermal conductivity along different directions and for different samples merely differs by a constant factor. With this assumption, we calculated the magnon contribution

$$\kappa_{m,a} = \kappa_a - K \cdot \kappa_b$$

with $K = 2.7$ and $K = 2.3$ for samples S1 and J1, respectively. The $K$ values were calculated as the averaged ratio $\kappa_a/\kappa_b$ in the temperature interval between about 20 and 35 K. The phonon contributions $K \cdot \kappa_b$ for samples S1 and J1 are shown by the broken lines in Fig. 2.

The resulting values of $\kappa_m(T)$ are presented in Fig. 3. The uncertainties caused by the subtraction of two experimentally measured curves, each with a relative error of about 0.5%, are also displayed in Fig. 3. Because of the relatively large uncertainties inherent to the evaluation procedure below about 40 K, it is impossible to draw any conclusions about $\kappa_m(T)$ in that temperature region. Therefore, only data at temperatures above 50 K were considered for the following analysis. In the accessible regime, $\kappa_m(T)$ first increases with increasing temperature but tends to pass through a maximum or to reach saturation at higher temperatures. The maximum absolute values of $\sim 1$ W m$^{-1}$ K$^{-1}$ for the magnon thermal con-
The low-temperature specific heat of κ and the results of numerical calculations at T = 0 are Δ0 = 0.41J and V = 2.49J. The energy diffusion constant DE(T), calculated for the single-crystalline sample S1 as outlined above and using Cs(T) as calculated by Jolicoeur and Golfinelli, is shown in the left panel of Fig. 4; the data for the second sample, not shown here, are very similar. The spin diffusion constant DS(T) from Ref. 18 is also displayed in the same figure. The absolute values of the two parameters are similar in magnitude at high temperatures and they exhibit very similarly shaped temperature dependences. This is true although, in comparison with the corresponding feature of DS(T), the maximum value of DE(T) is about two times larger and slightly shifted to lower temperatures. The only other spin-chain compound for which data on both the spin and the energy transport are available, is the Heisenberg S = 1/2 spin chain compound Sr2CuO3. In the right panel of Fig. 4, we present both DS(T) data from Ref. 40 and DE(T) data calculated from results of our previous thermal conductivity measurements on Sr2CuO3. It is again obvious that both quantities vary similarly with temperature and DE ∼ 2DS in the covered temperature interval. The only other material for which DE and DS have been compared up to now is solid 3He in the region 0.05 < T < 0.12 K, where the ratio DE/DS ≈ 2.1 is, again, close to two. It is remarkable that indeed the factor of 2 difference between DE and DS is expected to be a general property of any Bravais lattice with identical spins on each lattice point. The fair agreement of our results with this expectation thus confirms the reliability of our calculation of the spin-mediated thermal conductivity.

The limited accessible temperature interval for the evaluation of km(T) and the increasing uncertainty with decreasing temperatures do not allow a serious analysis of its overall temperature dependence. The classical high-temperature limit of DE for a spin system is given by

\[ \kappa_m \propto D_E \propto T^{\frac{1}{2}} \]

FIG. 3: Magnon thermal conductivity along the a-axis of AgVP2S6, extracted from \( \kappa_m(T) \) for the two samples S1 (a) and J1 (b).

FIG. 4: The energy diffusion constant \( D_E(T) \), calculated from the thermal conductivity data of sample S1 (open circles), and the spin diffusion constant \( D_S(T) \) from Ref. 18 (solid circles) are shown in the left panel. In the right panel, the corresponding data for the \( S = 1/2 \) spin chain compound Sr2CuO3 are shown.
For AgVP$_2$S$_6$, this high-temperature limit is $D_{E,ht} \sim 10^{14}$ sec$^{-1}$. The observed values in the temperature range covered in this study are, except at the lowest temperatures, consistently higher than $D_{E,ht}$, but tend to approach this value with increasing temperature. The enhancement is, however, much less pronounced than in $S = 1/2$ chain and ladder compounds. A better way to demonstrate this is by invoking the average mean free path of itinerant spin excitations $\ell_m(T)$, which can be defined as $\ell_m \equiv D_E/v_m$, where $v_m(T)$ is the average group velocity

$$v_m(T) = \frac{1}{\hbar} \left[ \int \frac{\partial \varepsilon}{\partial k} f(\varepsilon, T) dk \right] / \left[ \int f(\varepsilon, T) dk \right].$$

The calculated values of $\ell_m(T)$, shown in the inset of Fig. 4, do not exceed 60 Å. In contrast, the mean free paths of spin excitations in many $S = 1/2$ chains typically reach values of the order of $10^3$ Å. In Ref. 49, where this approach was applied to S=1 HAFM chains, it is noted that this is true only in the case of a large and energy-independent $\tau$ or, equivalently, to a long mean free path $\ell$. However, impurity scattering, for example, is shown to invoke energy-dependent lifetimes, thus leading to a more complicated behaviour of $\kappa_m$ which is not simply given by a product of the thermal Drude weight and the average relaxation time. Since in our case, as reflected in the small values of $\ell_m$ which approach the interspin distance (see the inset of Fig. 4), the relaxation is quite strong, we do not attempt a direct application of the thermal Drude weight formalism to our result for $\kappa_m(T)$ of AgVP$_2$S$_6$. This approach seems, however, well justified for the analysis of $\kappa_s$ in $S = 1/2$ 1D spin systems, where the spinon mean free paths are often rather large.\textsuperscript{37}

![Image](5)

For an ideal integrable system, $\kappa_{\text{spin}}$ diverges if $D_{th}$ is nonzero. If perturbations introduce a finite lifetime $\tau$, the thermal conductivity is also expected to be finite and can be represented as $\kappa_{\text{spin}} = D_{th}\tau$. In Ref. 49, where this approach was applied to S=1 HAFM chains, it is noted that this is true only in the case of a large and energy-independent $\tau$ or, equivalently, to a long mean free path $\ell$. However, impurity scattering, for example, is shown to invoke energy-dependent lifetimes, thus leading to a more complicated behaviour of $\kappa_m$ which is not simply given by a product of the thermal Drude weight and the average relaxation time. Since in our case, as reflected in the small values of $\ell_m$ which approach the interspin distance (see the inset of Fig. 4), the relaxation is quite strong, we do not attempt a direct application of the thermal Drude weight formalism to our result for $\kappa_m(T)$ of AgVP$_2$S$_6$. This approach seems, however, well justified for the analysis of $\kappa_s$ in $S = 1/2$ 1D spin systems, where the spinon mean free paths are often rather large.\textsuperscript{37}

### IV. SUMMARY AND CONCLUSIONS

In conclusion, by probing the anisotropy of the thermal conductivity of the S=1 Haldane-gap HAFM compound AgVP$_2$S$_6$, we established the magnitude and the temperature variation of the spin-related energy transport in the temperature region between about 50 and 170 K.
By comparing the energy and the spin diffusion parameters $D_E(T)$ and $D_S(T)$, both derived from experimental data, we note that they not only exhibit similar temperature dependences but that also their absolute values are of the same order of magnitude. We argue that the previous suggestion of an intrinsic origin of the spin diffusion at high temperatures in this system also applies to the energy transport that is investigated here. The character of the energy transport at low temperatures remains to be investigated.

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