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Heat transport in real spin-chain materials

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Abstract

The problem of heat transport in low-dimensional antiferromagnets that consist of 1D spin-chains coupled to three-dimensional phonons and impurities is discussed. Using an example of SrCuO₂ doped with Ca we show that the thermal conductivity in this class of materials should exhibit a ubiquitous behavior—saturation at higher temperatures. The reasons for such a behavior are outlined and a close agreement with the existing experimental data is demonstrated.

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1. Introduction

Low-dimensional antiferromagnets show large thermal conductivity due to spin excitations [1,2]. A subgroup of these materials can be closely described by a model of 1D Heisenberg spin chains with the superexchange J that is much larger than the phonon Debye energy, Θ_D .

In our previous works [3] we have developed a microscopic approach to the problem of 1D spin excitations in spin chains coupled to 3D phonon environment and impurities. We have considered the spin heat transport within the Boltzmann equation framework with a particular focus on one of such spin-chain systems, Sr₂CuO₃. Perhaps the most dramatic prediction of our theory is the saturation of the thermal conductivity at $T > \tilde{\Theta}_D \approx \Theta_D/4$, the temperature of order 100–200 K.

Very recently, experiments have been carried out to separate the phonon part of the thermal conductivity κ_{ph} from the spin part κ_s in a controlled way [4]. This was done by introducing Ca in SrCuO₂, another spin-chain system whose spin excitations are very similar to the ones in Sr₂CuO₃. The Ca-doping was used to suppress the phonon thermal conductivity while leaving the spin part intact. The spin component of the thermal conductivity in

Sr_{0.95}Ca_{0.05}CuO₂ was found to be essentially flat at $T > 100$ K with the saturated value equal to the high- T value of κ_s in the undoped SrCuO₂. Furthermore, the broad peak at intermediate T , present in SrCuO₂ and Sr₂CuO₃ (see Fig. 1), is wiped out in the doped system.

In this paper we provide an explanation for both the saturation and the wipeout of the peak.

2. Spin thermal conductivity

We have shown [3] that the spin thermal conductivity in spin-chain materials is determined by impurity scattering at low T and by phonon and impurity scatterings at high T :

$$\kappa_s(T) \propto \begin{cases} T^2 & \text{for } T \ll T_m, \\ T^{-1} & \text{for } T_m \ll T \ll \tilde{\Theta}_D, \\ T^0 & \text{for } T \gg \tilde{\Theta}_D, \end{cases} \quad (1)$$

where $\tilde{\Theta}_D \approx \Theta_D/4$ and T_m is a crossover temperature between impurity- and phonon-scattering dominated regimes. [3]. In the case $T_m \ll \tilde{\Theta}_D$, T_m also corresponds to a maximum in $\kappa_s(T)$, see Fig. 1.

We predict a remarkable behavior of the thermal conductivity at the temperatures $T > \tilde{\Theta}_D$: saturation at a constant value. A parallel can be drawn with the thermal conductivity of metals, where κ saturates at $T \sim \Theta_D$ where the phonon scattering dominates. However, the reason for

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Fig. 1. (From Ref. [3]). Normalized spin thermal conductivity κ_s vs reduced temperature T/T_m . Diamonds are the experimental data for Sr_2CuO_3 , $T_m = 79.4\text{ K}$ and $\kappa_s^{\text{max}} = 36.7\text{ W m}^{-1}\text{ K}^{-1}$, see Ref. [2]. Shaded area schematically represents the range where the phonon background subtraction creates a large uncertainty in the data [2]. Solid line is the result of this work, see Ref. [3], for $\Theta_D = 11.6T_m$. Arrows mark the saturation value κ_s^∞ for the solid line at $T \gg \tilde{\Theta}_D$ and the spin-phonon scattering crossover scale $\tilde{\Theta}_D$, respectively. Asymptotic behavior at $T \gg T_m$ ($\sim \text{const}$) is shown by the dashed line.

Fig. 2. Spin thermal conductivity κ_s vs temperature T . Diamonds are the experimental data for $\text{Sr}_{0.95}\text{Ca}_{0.05}\text{CuO}_2$ and $T_m = 58\text{ K}$ see Ref. [4]. Notations are the same as in Fig. 1.

the saturation in spin chains is quite subtle. It comes about as a combination of T - and k -dependencies of the impurity and phonon relaxation rates and the 1D density of states of spin excitations that are universal in the long-wavelength limit. The major contribution to the spin thermal current in spin chains comes from the long-wavelength excitations, $k \ll T/J$. As the temperature increases, this enhances phonon scattering and shrinks the relevant energy region for spin excitations. Therefore, our long-wavelength results become more precise at higher temperatures.

In our theory spin excitations pass the non-equilibrium momentum to the phonon system that dissipates it via an Umklapp or impurity scattering. Thus, the phonon system is treated as a thermal bath. This requires the phonons to be sufficiently equilibrated, or $\tau_p \ll \tau_s$, where τ_s and τ_p are

the spin and phonon relaxation times, respectively. This inequality holds very well for $T > \tilde{\Theta}_D$ because of the frequent phonon–phonon scattering, but it can be violated as T decreases below $\tilde{\Theta}_D$ for the pure systems.

On the other hand, lattice disorder due to Ca doping would facilitate $\tau_p \ll \tau_s$ for all temperatures because impurities allow phonons to dissipate their momentum effectively. Thus, equilibration of phonons in doped systems becomes fast even at lower temperatures. In that case, our theory should work very well. This is demonstrated in our Fig. 2. The thermal conductivity of the 5% Ca-doped SrCuO_2 exhibits a saturation at $T > \tilde{\Theta}_D$ in an excellent agreement with our result for $\Theta_D = 400\text{ K}$.

We note that the theoretical κ_s from Ref. [3] for the pure Sr_2CuO_3 also agrees well with the experimental data, Fig. 1, but it requires a rather large Debye energy $\Theta_D \approx 950\text{ K}$ to fit the maximum. Moreover, for the undoped SrCuO_2 we could reach only a qualitative agreement with the data and that also required an unphysically large Θ_D . If, on the other hand, we select a reasonable $\Theta_D = 400\text{ K}$ we obtain the curve shown in our Fig. 2.

3. Spin-phonon drag

We have: (a) κ_s is flat when the relaxation of phonons is fast due to doping, and (b) with no doping κ_s has a maximum at lower T where phonon relaxation may not be fast. Then, what is the origin of the maximum? We propose the answer: spin-phonon drag. That is, spin excitations pass on a non-equilibrium momentum to the phonon system and thus increase the phonon thermal current. In other words, in the undoped systems there is an interference effect: κ_{total} is not simply $\kappa_s + \kappa_{\text{ph}}$. The spin-phonon drag is insignificant at high T because the phonon relaxation is effective, but it becomes significant at low T . When the lattice is sufficiently doped, the internal relaxation in the phonon system becomes very effective at all temperatures and the drag is suppressed. To verify this suggestion, a systematic experimental study of the doping-dependence of the thermal conductivity in spin-chain systems is necessary. To calculate the spin-phonon drag effect using our approach, one needs to solve a system of Boltzmann equations for spin excitations and phonons without an assumption of the equilibrium distribution of phonons.

4. Summary

We have explained a prominent feature of the thermal conductivity in spin-chain materials—saturation at high temperatures. We propose that the increase in κ_s at low temperatures in the undoped systems should be due to spin-phonon drag. Impurity doping leaves the spin heat current intact but suppresses the spin-phonon drag by limiting the phonon relaxation time. Further experiments are suggested and future directions for the theoretical work are outlined.

Acknowledgments

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