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Thermal conductivity of some soliton-bearing magnetic systems

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The thermal conductivity λ of $[C_6H_{11}NH_3]CuBr_3$ (CHAB) has been measured in the region 1.5 K $<T<$ 10 K and $B<7$ T with the heat flow parallel to the chain direction. The data on this soliton-bearing system, as well as earlier data on $[CH_3]_4NMnCl_3$ (TMMC), are compared with the results of a model describing the heat conduction by magnons and solitons that was proposed recently by Wysin and Kumar. The observed qualitative behavior of λ_B/λ_0 in both systems systematically deviates from the corresponding prediction, which is most likely due to the fact that in the model the heat transport by the photon system is neglected.

The suggestion that the dominant nonlinear excitations in certain classes of one-dimensional magnetic systems can be associated with sine-Gordon solitons has prompted a large theoretical and experimental effort.[1-3] Up till now, however, hardly any attention has been given to the effect of solitons on the thermomagnetic transport properties of such systems. Recently, Wysin and Kumar[4] (hereafter referred to as WK) proposed a model in which these effects were taken into account. In this Rapid Communication their results will be confronted with the experimentally determined thermal conductivity of two well-known soliton-bearing systems, the $S=\frac{1}{2}$ ferromagnetic chain system $[C_6H_{11}NH_3]CuBr_3$ (CHAB)[5,6] and the $S=\frac{5}{2}$ antiferromagnetic chain system $[CH_3]_4NMnCl_3$ (TMMC).[2,7]

The thermal conductivity λ of CHAB was measured[8] by a steady-state longitudinal heat-flow method. Using a two-step measuring technique,[8] temperature differences as small as 0.1 mK could be determined with an inaccuracy less than 2%. The performance of the setup was tested by means of a reference sample consisting of stainless steel (SRM 735).[9] The difference between the measured conductivity and the reference values[9] given by the National Bureau of Standards was less than 2%, which agrees with the reported inaccuracy. The experiments were performed in the range 1.5 K $<T<$ 10 K with the heat flow chosen parallel to the chain direction and a magnetic field ($B<7$ T) applied along the a, b, or c axis. A typical set of data collected with the magnetic field applied parallel to the c axis (which is located in the easy plane, allowing the presence of solitonlike excitations) is shown in Fig. 1. This figure reveals that the normalized thermal conductivity λ_B/λ_0 systematically increases with increasing field. In the course of the experiments we observed that the absolute magnitude of λ decreased upon thermal cycling between 4.2 K and room temperature, probably due to an increase of lattice imperfections. Although the qualitative behavior of λ_B/λ_0 was not affected, we have included in Fig. 1 only data obtained from experiments on a fresh single crystal after the first cool down. The zero-field thermal conductivity of this crystal is shown in Fig. 2(a).

Now we will turn to a comparison of our data with the results of the model proposed in Ref. 4. We will briefly summarize the most relevant characteristics of this model, which describes the thermal conductivity associated with the magnetic degrees of freedom in a one-dimensional system. In the model, the concept of a local equilibrium distribution[10] is used together with the linearized Boltzmann equation in the relaxation-time approximation. It is assumed that all interactions between the magnetic excitations can be ignored, except for a weak soliton-magnon interaction, which is taken into account by a modification of the soliton equilibrium distribution. The relaxation towards equilibrium is attributed to an extrinsic mechanism, e.g., magnetoelastic coupling. It is assumed that this coupling does not affect the dispersion relation and the distribution function of magnons and solitons itself. The extrinsic relaxation rate is assumed to be dominant in the sense that relaxation due to the soliton-magnon scattering can be neglected. Within this framework the contribution of solitons and magnons to the thermal conductivity, denoted by λ_{sol} and λ_{mag}, respectively, can be added linearly, and are given by

$$\lambda_{sol}=\frac{\tau_{sol}c_0}{\beta\sqrt{2\pi}}4\epsilon_0(\beta E_0)^{-1/2}e^{-\beta E_0}[(\beta E_0)^2+\beta E_0+\tfrac{7}{4}] \;, \tag{1}$$

$$\lambda_{mag}=\frac{\tau_{mag}c_0}{\beta^2}(\beta\epsilon_0)^{-1}[S_2(\beta\epsilon_0)] \;, \tag{2}$$

with

$$S_2(x_0)=\frac{1}{4\pi}\int_{x_0}^{\infty}dx\frac{(x^2-x_0^2)^{1/2}}{\sinh^2(\frac{1}{2}x)} \;.$$

Here $\tau_{mag(sol)}$ represents the relaxation time of the magnons (solitons) and $\beta=1/k_BT$. E_0 is the soliton rest energy, ϵ_0 is the magnon gap, and c_0 is the magnon velocity. Formally these equations are valid for both ferromagnetic

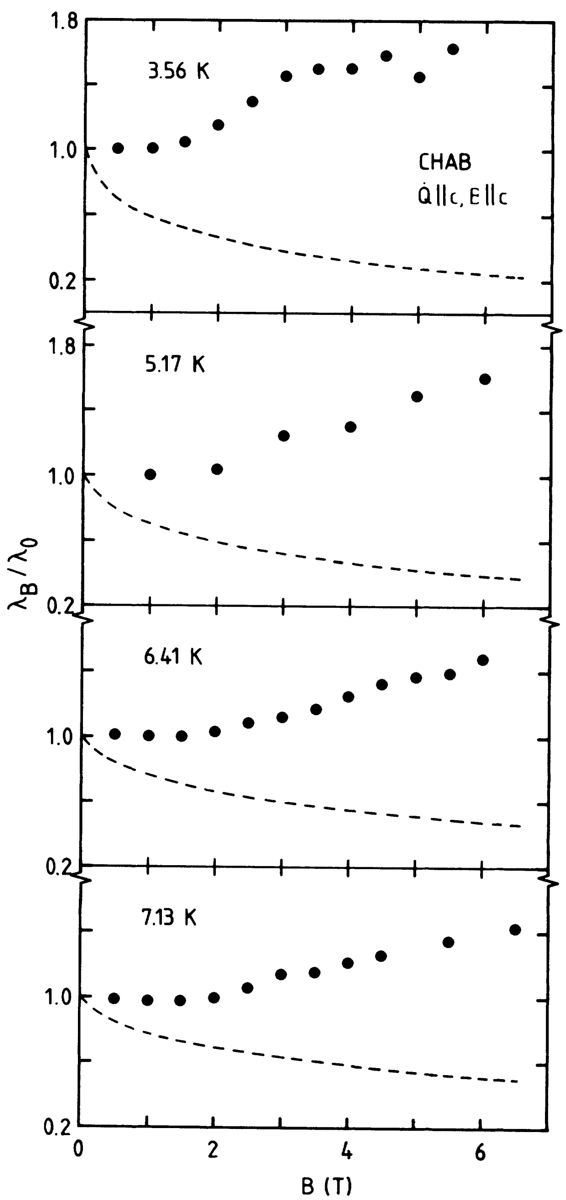

FIG. 1. Isothermal field dependence of the thermal conductivity λ in CHAB with the magnetic field (B) applied along the c axis. The data are normalized with respect to the zero-field value λ_0. The dashed lines are calculations based on the theory of WK with $\tau_{\rm sol} = \tau_{\rm mag}$.

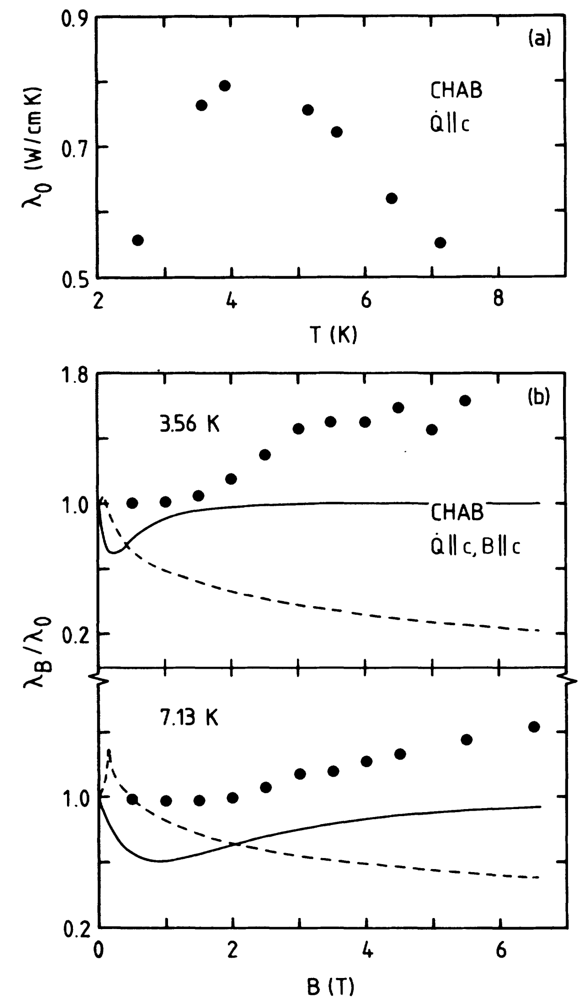

FIG. 2 (a) Zero-field thermal conductivity λ_0 for CHAB as function of the temperature for a fresh single crystal. (b) Calculated field dependence of the normalized thermal conductivity λ_B/λ_0 in CHAB based on the theory of WK with $\tau_{\rm sol} = 10\tau_{\rm mag}$ (dashed curves) and the resonant soliton-phonon interaction model (solid curves). In the last case the calculations were performed for the same ratio of magnetic and nonmagnetic scattering as in the case of TMMC (Figs. 3 and 4). The black circles represent the data.

and antiferromagnetic systems.

If we insert the values appropriate to CHAB, i.e., $E_0/k_B = (34.3\ \mathrm{K\,T}^{-1/2})\sqrt{B}$ and $\epsilon_0 = (2.64\ \mathrm{K\,T}^{-1/2})\sqrt{B}$, in Eqs. (1) and (2), and assume the relaxation time of the solitons and magnons to be equal ($\tau_{\rm sol} = \tau_{\rm mag}$), we obtain a theoretical prediction that is reflected by the dashed curves in Fig. 1. Inspection of this figure reveals that, in contrast to the data, the predicted thermal conductivity λ_B/λ_0 monotonically decreases with increasing magnetic field. If we, intuitively, assume the relaxation of the solitons to be much slower than that of the magnons, and correspondingly take $\tau_{\rm sol} = 10\tau_{\rm mag}$, the high-field behavior of λ_B/λ_0 remains unaltered, but a maximum is predicted at fields below 0.5 T. This maximum, which in the model is associated with a soliton contribution, is not reflected by our data, as illustrated in more detail in Fig. 2(b). One should note, however, that in this field region classical soliton theory is formally no longer valid ($\beta E_0 > 5$),[11] and therefore any prediction involving a large soliton contribution in this region should be considered with some reservations.

Before we discuss a possible origin of the systematic qualitative discrepancy between theory and experiment, we will consider the measurements of the thermal conductivity of TMMC reported before.[12] The reason for this is twofold. First, to our knowledge these are the only available data on the thermal conductivity of a soliton-bearing system, apart from those presented above. Second, it seems interesting to compare the field dependence of λ of a ferromagnetic and an antiferromagnetic chain, since it is

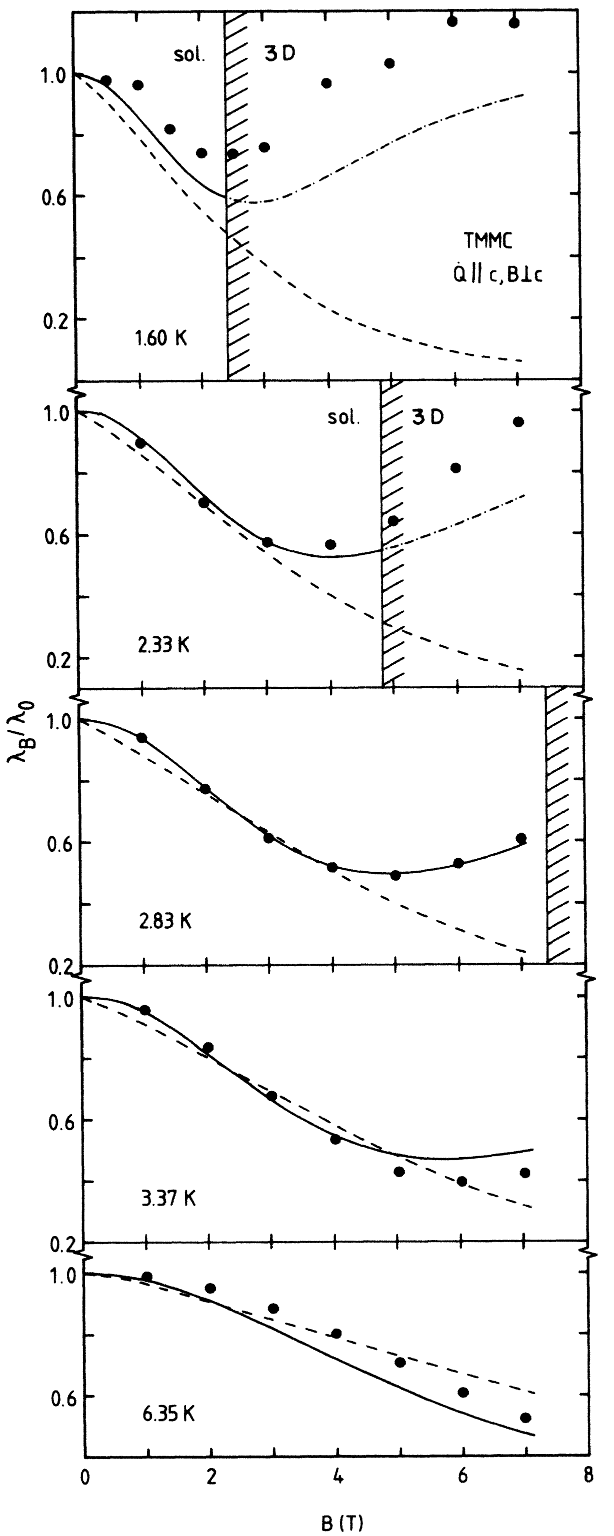

FIG. 3. Isothermal field dependence of the thermal conductivity λ in TMMC with the magnetic field B applied perpendicular to the c axis. The thermal conductivity is normalized with respect to the zero-field value λ_0. The solid and dashed curves represent calculations based on the resonant soliton-phonon interaction model and the theory of WK with $\tau_{sol} = \tau_{mag}$, respectively. The shaded areas indicate (the transition to) the 3D ordered state.

not *a priori* clear that the behavior should be similar in both cases.

For convenience, part of the experimental results for TMMC (Ref. 12) are reproduced in Fig. 3. It is obvious that the data on λ_B/λ_0 show a rather pronounced minimum, which shifts to higher fields when the temperature increases. This behavior is in marked contrast with the results for CHAB and was originally interpreted[12] using a model based on resonant soliton-phonon scattering (analogous to resonant magnon-phonon scattering). The basic idea behind this model is that the heat is transported by the phonon system, whereas the magnetic excitations effectively act as a scattering mechanism. If the dominant excitations in the spin system are magnons, this model predicted a gradual *increase* of λ at higher fields, which is directly related to the decrease of the magnon density. On the other hand, if the excitation spectrum is dominated by solitons, as is assumed for TMMC, the maximum in the soliton density occurring at finite values of B will give rise

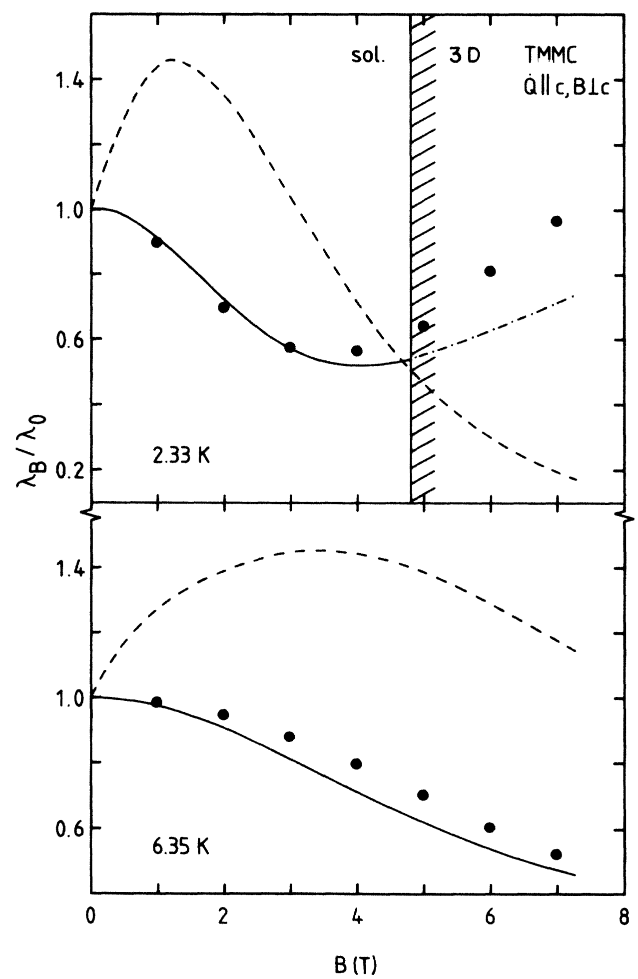

FIG. 4. Calculated field dependence of the normalized thermal conductivity λ_B/λ_0 in TMMC based on the resonant soliton-phonon coupling model (solid curve) and the theory of WK with $\tau_{sol} = 10\tau_{mag}$ (dashed curve). The shaded area indicates (the transition to) the 3D ordered state. The black circles represent the data.

to a minimum in λ_B/λ_0. Although the exact mechanism for soliton phonon scattering is not quite clear, the predicted behavior of λ_B/λ_0, reflected by the full curves in Fig. 3, is in very good agreement with the data, at least in the field and temperature range outside the three-dimensional (3D) ordered region. It is clear from the figure that within the ordered region systematic deviations occur. To some extent, these deviations support the interpretation of the field dependence of λ in terms of solitons, since the π solitons in an antiferromagnetic chain are strongly damped in the 3D ordered state.[7,13]

Alternatively, we have tried to describe the data on TMMC using the model proposed by Wysin and Kumar.[4] For this antiferromagnetic chain system the parameters E_0 and ϵ_0 in Eqs. (1) and (2) depend linearly on the applied magnetic field. Using the generally accepted parameter values for TMMC, i.e., $E_0/k_B=(2.6\ \mathrm{K}\,\mathrm{T}^{-1})B$ and $\epsilon_0/k_B=(1.04\ \mathrm{K}\,\mathrm{T}^{-1})B$, and assuming that $\tau_{\mathrm{sol}}=\tau_{\mathrm{mag}}$, we obtain a result that is reflected by the dashed curves in Fig. 3. Although this prediction agrees fairly well with the data collected at low fields, the experimentally observed minimum in λ_B/λ_0 is not reproduced. An increase of the relaxation time of the solitons up to $\tau_{\mathrm{sol}}=10\tau_{\mathrm{mag}}$ gives rise to large deviations from the experimental results, as is illustrated in Fig. 4 for two representative sets of data.

Now we return to the experiments on CHAB. Following the arguments given above, the gradual increase of the experimental data on λ_B/λ_0 with field indicates that in this compound the magnetic excitations also effectively act as a scattering mechanism. Since no clear minimum in λ_B/λ_0 is observed, the contribution of solitons seems to be rather small. Inspection of Fig. 1 already showed that the calculations based on the model proposed by Wysin and Kumar yield a decrease of λ_B/λ_0 at high fields, instead of the experimentally observed increase. Since the same qualitative tendency is observed in TMMC, we conclude that this discrepancy is most likely a consequence of the fact that in their model the transport of thermal energy by the phonon system itself is not taken into account.

Finally we would like to comment on the observation that the experimental data on CHAB and TMMC show a completely different behavior. Whereas in TMMC a satisfactory description of the data requires a significant contribution of solitons, such a large soliton contribution does not seem to be present in CHAB. It is not clear whether this difference is merely accidental or that it is related to the mechanism of the soliton-phonon scattering, which may be more effective for an antiferromagnetic chain system (TMMC) than for a ferromagnetic system (CHAB). Measurements on other soliton-bearing systems, e.g., $\mathrm{CsNiF_3}$, an $S=1$ ferromagnetic chain compound, may be necessary to obtain more information on this question.

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