Thermal conductivity of the ferromagnetic chain system $CsNiF_3$

H. A. M. de Gronckel, W. J. M. de Jonge, and K. Kopinga
Department of Physics, Eindhoven University of Technology, 5600-MB Eindhoven, The Netherlands

L. F. Lemmens
Institute for Applied Mathematics, University of Antwerp (RUCA), Groenenborgerlaan 171, B-2020 Antwerpen, Belgium

(Received 18 March 1991)

The thermal conductivity λ of $CsNiF_3$ has been measured in the region 2.5 K $< T <$ 10 K with the heat flow parallel to the chain direction. Fields up to 8 T were applied both within and perpendicular to the easy plane. The data on this ferromagnetic chain system indicate that the effect of solitons on λ is very small, in agreement with earlier observations on $[C_6H_{11}NH_3]CuBr_3$ (CHAB). This suggests that the soliton-phonon scattering in these systems is much smaller than in comparable antiferromagnetic systems, like tetramethylammonium manganese trichloride (TMMC).

A variety of theoretical and experimental studies[1-6] have shown that the effect of solitons on the static and dynamic properties of magnetic chain systems may be significant in a certain range of fields and temperatures. Relatively little attention, however, has been given to the thermomagnetic transport properties. One of our previous studies[7] revealed that in several experimental realizations of these systems heat is primarily transported by the phonons, whereas the magnetic excitations effectively act as a scattering mechanism. Moreover, it was found that in the antiferromagnetic chain system $[CH_3]_4NMnCl_3$ tetramethylammonium manganese trichloride (TMMC) a satisfactory description of the data required a significant contribution of solitons, whereas in the ferromagnetic chain system $[C_6H_{11}NH_3]CuBr_3$ such a large contribution was not observed. To investigate whether this results from a characteristic difference between ferro- and antiferromagnetic chain systems, probably related to a difference in soliton-phonon scattering, we have studied the thermal conductivity of the soliton-bearing system $CsNiF_3$.

The thermal conductivity λ of $CsNiF_3$ was measured 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). 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 2.5 K $< T <$ 10 K with the heat flow chosen parallel to the chain direction. The magnetic field (B $<$ 8 T) was applied within the easy plane ($\mathbf{B}\perp c$), allowing the presence of solitonlike excitations, or perpendicular to it ($\mathbf{B}||c$). During the measurements it appeared that the single crystals rapidly deteriorated upon thermal cycling between room temperature and 4.2 K, which manifested itself in a significant decrease of the magnitude of λ. In this paper we will confine ourselves to results obtained on fresh single crystals after the first cool down. In Fig. 1 we have plotted the temperature dependence of λ at zero field. The data show a scatter of about 10%, which is due to the fact that the $CsNiF_3$ crystals were very fragile; this fragility prevented us from creating good thermal contact between heater, sample, and heat sink. Hence only very small temperature gradients within the crystals could be achieved.

We will now turn to the field dependence of λ. In Fig. 2 we have plotted the observed variation of the normalized thermal conductivity λ_B/λ_0 with B at various constant temperatures and the field B applied within the easy plane. The data show a gradual increase of λ_B/λ_0 of about a factor 2 with increasing field, whereas hardly any field dependence is observed above $B=3$ T. For comparison we have included in the figure two different theoretical predictions. First, we have used the model proposed by Wysin and Kumar[10], that we also applied in the analysis of the thermal conductivity of $[C_6H_{11}NH_3]CuBr_3$

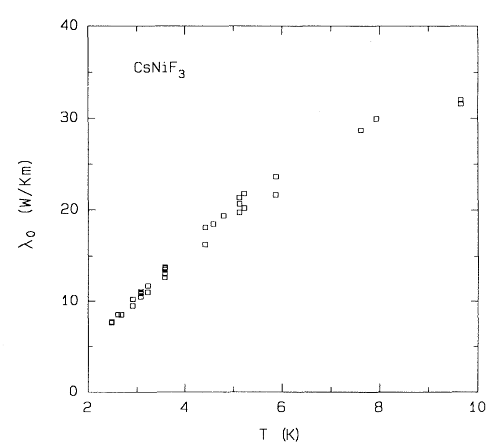

FIG. 1. Zero-field thermal conductivity λ_0 for $CsNiF_3$ as a function of temperature for a fresh single crystal.

and TMMC. Inspection of Fig. 2 reveals that the resulting prediction for λ_B/λ_0, which is represented by the dashed-dotted curves, *decreases* at higher fields, in contrast to the data. As already argued in a previous study,[7] this discrepancy most likely results from the fact that in this model the heat transport by the phonon system is not taken into account. Qualitatively, the data are consistent with a situation where the heat is mainly transported by the phonons, whereas the magnetic excitations, which are suppressed at high fields, act as an effective scattering mechanism. A complete theoretical description of such a situation is —at present— not possible. However, the main features of the observed behavior can be described correctly by a simplified model based on resonant scattering between the phonons and the magnetic excitations[11]. Within this model, the thermal conductivity λ is given by

$$\lambda=C_m T^m \int_{x_0}^{x_{\max}} \frac{x^{m+1}e^x}{(e^x-1)^2}\left[\tau_{\rm ph}^{-1}+\sum_R\left[\tau_R^{-1}(n_R/n_{\rm ph})\frac{x^2}{(x-x_{R,0})^2}\right]\right]^{-1} dx \ . \tag{1}$$

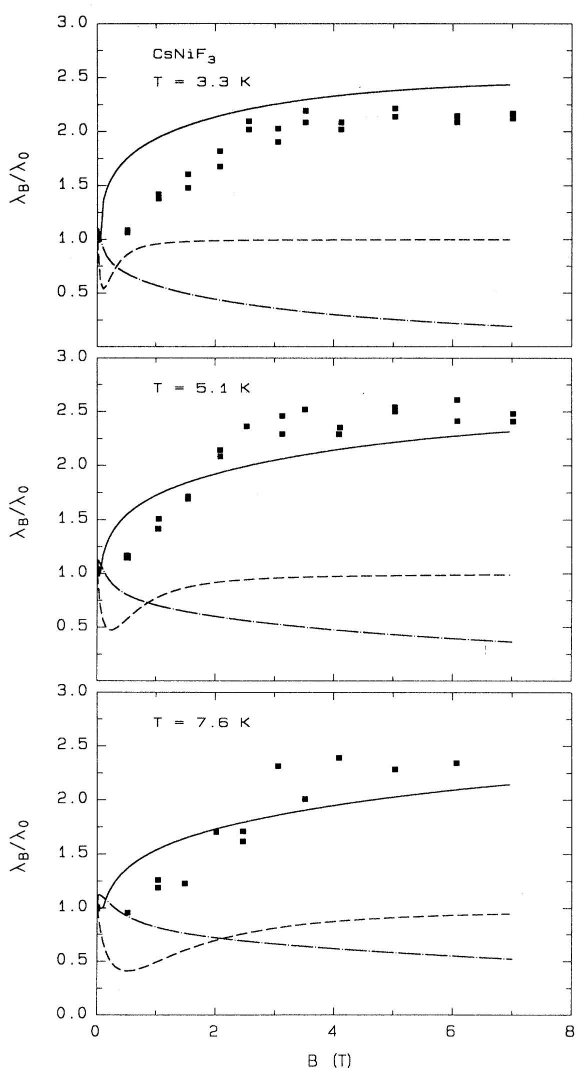

FIG. 2. Isothermal field dependence of the thermal conductivity λ in $CsNiF_3$ with the magnetic field B applied perpendicular to the c axis. The thermal conductivity is normalized with respect to the zero-field value λ_0. The dashed-dotted curves represent calculations based on the model of Ref. 10 with $\tau_{\rm sol}=\tau_{\rm mag}$. Calculations based on the resonant-interaction model (Ref.11) are reflected by dashed and solid curves, which represent the bare effect of solitons and magnons, respectively.

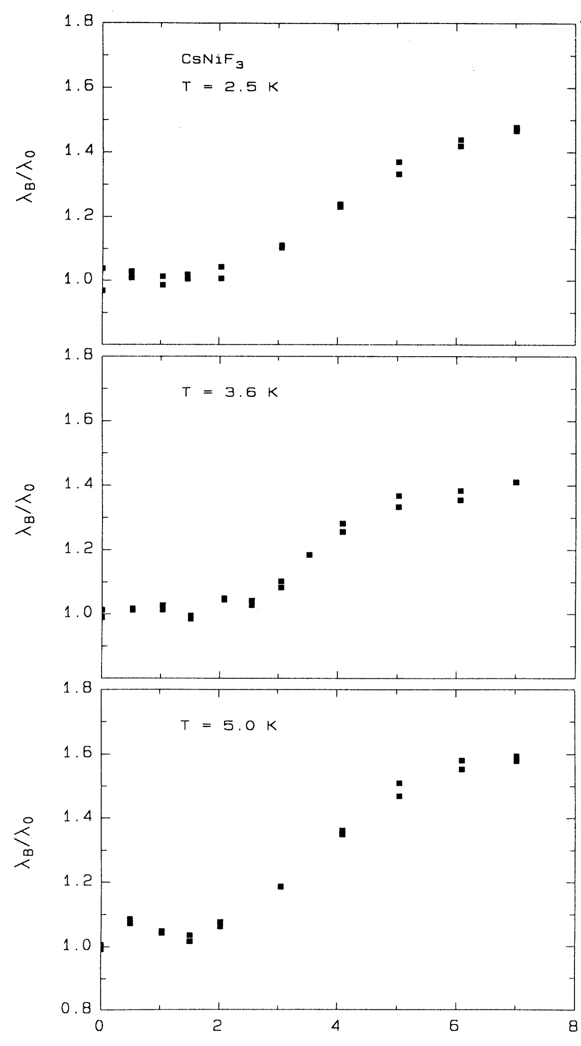

FIG. 3. Isothermal field dependence of the thermal conductivity λ in $CsNiF_3$ with the magnetic field B applied along the c axis. The data are normalized with respect to the zero-field value λ_0.

In this equation C_m is a proportionality factor, m the effective dimensionality, $x = \varepsilon / k_B T$ with ε the energy, τ_{ph} is the relaxation time of the phonon system, and τ_R represents the relaxation due to magnons and solitons, respectively. The parameter x_{max} is the usual Debye cutoff, $x_{R,0}$ represents the energy of the respective magnetic excitations at zero wave number, and n is the number operator. Note that in the calculation of λ_B / λ_0 the prefactor $C_m T^M$ cancels out.

If the dominant magnetic excitations are magnons, this model predicts a gradual increase of λ_B / λ_0 at increasing B. If, on the other hand, the scattering of the phonons is mainly caused by solitons, a minimum in λ_B / λ_0 is predicted at the field where the soliton density n_{sol} has a maximum, i.e., $B = 9.97 \times 10^{-4}$ T^2 for $CsNiF_3$. In the actual calculations we used the data collected at $T = 7.6$ K to normalize the ratio of the magnetic to the nonmagnetic scattering. In Fig. 2 we have plotted the calculated variation of λ_B / λ_0 with B resulting from solitons (dashed curves) or magnons (solid curves). The effective dimensionality m of the system was chosen equal to one. Inspection of this figure reveals that, despite the obvious simplicity of the model, the main features of the experimental data are reproduced correctly if only magnon scattering is taken into account. This strongly suggests that the effect of solitons on λ_B / λ_0—if actually present—is of minor importance.

We will now consider the measurements for fields perpendicular to the easy plane ($\mathbf{B} \perp c$), which are plotted in a normalized way in Fig. 3. The general characteristics of the data are an almost-field-independent behavior below $B = 2$ T, a gradual increase for 2 T $< B <$ 6 T, and saturation at higher fields. If we assume for this direction of **B** the dominant excitations to be magnons, this behavior can qualitatively be explained as follows. At low fields perpendicular to the easy plane, the ground state of the system is highly degenerate (Goldstone mode). This degeneracy is lifted at fields that are high enough to overcome the easy-plane anisotropy. Within a classical (mean-field) approach, this field is given by the equation $g \mu_B B S = D S^2$. Inserting the values appropriate to $CsNiF_3$, i.e., $g = 2.1$, $S = 1$, and $D / k_B = 4.5$ K (classical[2]), the magnon density is predicted to start to decrease at $B = 3$ T. This agrees surprisingly well with the observed increase of λ_B / λ_0, indicating that also for this direction of **B** the picture in which the heat is mainly carried by the phonons, which are effectively scattered by the magnons, is appropriate.

Concluding we like to state that the present study, together with earlier results on $[C_6H_{11}NH_3]CuBr_3$ and TMMC[7], strongly indicates that the soliton-phonon scattering in ferromagnetic chain systems is much less effective than in their antiferromagnetic counterparts. Probably this difference is related to the difference between the dynamic structure factors[1-3] of the solitons in these two types of systems.

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