

T diagram depends on the magnitudes of ΔT and \mathcal{K}_L ; in particular, the Lifshitz field \mathcal{K}_L should be accessible. According to Ref. 5, $\Delta T \sim 1^\circ\text{C}$. Unfortunately, \mathcal{K}_L cannot be estimated so long as the magnitude of α in (12) is unknown. However, \mathcal{K}_L can be made arbitrarily small by preparing a sufficiently dilute solution of the chiral material in question in a nonchiral solvent. Indeed, $K_2 \rightarrow 0$ when the concentration of the chiral solute goes to zero.⁷ Then, according to (12) also $\mathcal{K}_L \rightarrow 0$. Another method to decrease \mathcal{K}_L is to prepare a mixture of two isomers of the same chiral molecule and to decrease the difference of their concentrations. Since $\Delta T \propto \mathcal{K}_L^2$, the required decrease of \mathcal{K}_L to an available magnitude may (*not necessarily*) diminish ΔT so as to make it difficult to see the details of the phase diagram (Fig. 1 will undergo a strong contraction in the vertical direction). However, since the point (\mathcal{K}_L, T_L) itself *can* be reached, it is possible to test the predictions of Ref. 1 concerning the critical behavior near this point.

Although the fluctuation corrections to the Landau theory affect the values of critical exponents, I expect that the existence of the Lifshitz point established above will be unaffected. Indeed, if one applies the renormalization-group method of Wilson,¹⁰ one usually starts from the Landau free-energy functional (the Landau-Wilson Ham-

iltonian). The procedure of finding the actual symmetry-breaking order parameter (the "soft-mode") of the phase transition is then the same in the Landau theory.¹¹ Since *only* this procedure has been used above to establish the existence of the Lifshitz point, the result should not be affected by the renormalization.

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Energy Transport above T_c by Paramagnetic Magnons in Two-Dimensional Ferromagnetic Heisenberg Systems

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The thermal conductivity of the two-dimensional magnetic systems $(C_nH_{2n+1}NH_3)_2CuCl_4$ (with $n=1,2$) has been measured from $0.12T_c$ to $2.5T_c$ in fields up to 6.5 T. We found a considerable heat transport by the magnetic system, both below and above T_c . Above T_c this heat transport can be attributed to magnonlike states which can exist well into the paramagnetic region in low-dimensionality systems.

The dynamics of fluctuations in both ordered and disordered spin systems are of considerable current interest. The existence of these spin fluctuations at temperatures far above the critical temperature T_c has been established by neutron scattering experiments for three-dimensional systems¹ as well as for systems of lower dimensionality.^{2,3} But their functioning as agents of energy transport has never been observed before. In this Letter we report for the first time experi-

mental evidence of the transport of thermal energy in the paramagnetic region by the spin system. This evidence follows from thermal-conductivity measurements in two-dimensional (2D) Heisenberg systems which show in the ordered phase a fractional magnon contribution of 90%—the largest yet observed—and above T_c a fractional magnetic contribution to the heat transport, ranging from 86% at T_c to a maximum of 95% at $T=1.5T_c$, which can mainly be attributed to paramagnetic

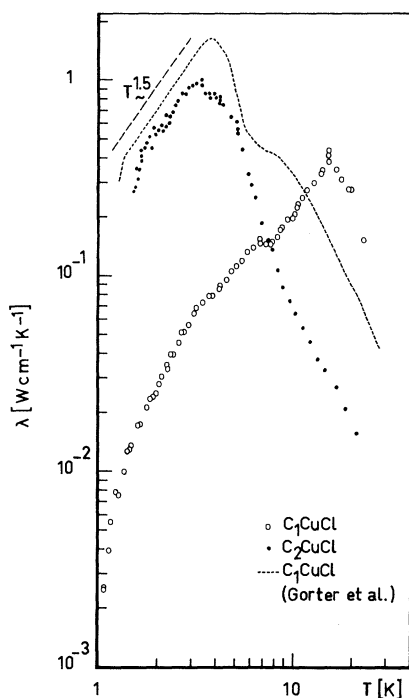


FIG. 1. Temperature dependence of the thermal conductivity of C_1CuCl and C_2CuCl at zero field. The broken line is proportional to $T^{1.5}$.

magnons.

The thermal-conductivity experiments were performed on two copper compounds of the general formula $(C_nH_{2n+1}NH_3)_2CuCl_4$, with $n = 1, 2, \dots$ (henceforth abbreviated to C_nCuCl), which consist of sheets of nearly quadratically arranged Cu^{2+} ions, widely separated by a double layer of $(C_nH_{2n+1}NH_3)$ chains.⁴ Since the intralayer interaction J is far stronger than the interlayer interaction $J' = RJ$ (with $|R| = 10^{-3} - 10^{-6}$), these crystals approximate a 2D, $S = \frac{1}{2}$ Heisenberg ferromagnet to a very high degree. Besides exhibiting a 2D magnetic structure, it was shown that the phonon system is 2D in character as well.^{5,6} We have measured the thermal conductivity of C_1CuCl ($T_c = 8.895$ K) and C_2CuCl ($T_c = 10.20$ K) in the temperature range between 1 and 23 K, and in fields up to 6.5 T. Figure 1 shows the zero-field results of both crystals. For temperatures below 3 K, the C_1CuCl values are about an order of magnitude smaller than the C_2CuCl results. This is due to the lesser quality of the C_1CuCl crystal. On examination after the measurements it was found that the C_1CuCl sample showed some internal cracks, while the C_2CuCl crystal was still of excellent quality. This quality difference was reflected by the estimated dislocation densities,

which for the C_1CuCl crystal was about two orders of magnitude higher than for the C_2CuCl sample.⁶ For comparison purposes, the zero-field results obtained by Gorter *et al.*⁷ for a good-quality C_1CuCl crystal are also given in Fig. 1. Below 3 K, the C_2CuCl sample shows a $T^{1.5}$ behavior, indicative of a dominating 2D magnon conduction.⁷

To separate the magnon from the phonon contribution the following procedure was used. For $T < T_c$, the magnon contribution in both crystals could be completely quenched for fields of 6 T, and the resultant conductivity could be taken as the lattice conductivity of the crystals. For $T > T_c$, it appeared that in the case of C_1CuCl the thermal conductivity as a function of field did not yet reach a saturation value at fields of 6.5 T and the magnon contribution was still not completely suppressed at that high field value. To estimate the phonon contribution for $T > T_c$, we used the same procedure as employed by Gorter *et al.*,⁷ which consists of equating the lattice conductivity to the properly scaled thermal conductivity of the diamagnetic, isomorphous C_1CdCl compound which has phonon conduction only. The following results were obtained: Below T_c the fractional decrease due to the field is about 75% for the C_2CuCl crystal and about 90% for the C_1CuCl sample (Fig. 2). This is the largest fractional magnon contribution to the energy transport yet measured. It should also be noted that the good quality C_2CuCl sample follows the prediction for a 2D noninteracting magnon gas far better than the C_1CuCl crystal which shows a steeper decrease with field (Fig. 2).

Above T_c for the C_1CuCl crystal we still found a very large contribution of the magnetic system, not only in zero field but in the largest fields as well (Fig. 3). As also can be seen from Fig. 1, the thermal conductivity of the C_1CuCl crystal at zero field, apart from a slight dip somewhat below T_c , rises monotonically to a maximum at 15.5 K, and even exceeds the zero-field values of the good-quality C_1CuCl crystal. The observed thermal-conductivity maximum coincides with the maximum of the magnetic specific heat, as measured by Bloembergen.⁸ This is in marked contrast to the zero-field behavior of the good-quality C_1CuCl and C_2CuCl crystals, which show a decrease above 3.5 K and for which the non-zero-field values coincide with the zero-field values above T_c .^{7,9}

The existence of magnonlike excitations well into the paramagnetic region and even in the ab-

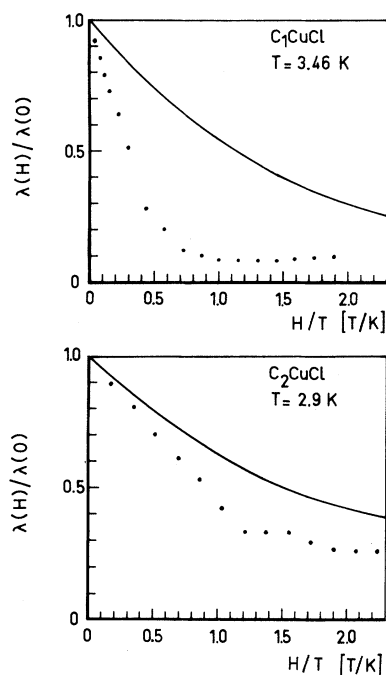


FIG. 2. Field dependence of the thermal conductivity of C_1CuCl and C_2CuCl . The solid line represents the sum of the conductivity of the theoretically calculated noninteracting magnon gas and the estimated lattice conductivity.

sence of long-range order has first been explained by Marshall.¹⁰ In systems of lower dimensionality, the magnon modes are even more pronounced above T_c , as is for instance exhibited in the 2D K_2NiF_4 ² and the 1D TMMC $[(CD_3)_4NMnCl_3]$.³ No theory for the energy transport by these paramagnetic magnons exists as yet, however. We have also calculated^{11,12} the possible contribution of long-wavelength diffusive spin modes to the thermal conductivity, but this does not exceed 5% of the observed λ . For that reason we have attributed the magnetic thermal conduction above T_c as mainly due to paramagnetic magnons.

The question then remains why the good-quality C_1CuCl and C_2CuCl crystals do not show a large magnetic contribution above T_c . A clue for this difference in behavior above T_c might be found in their behavior below T_c . As can be seen from Fig. 2, the good-quality C_2CuCl crystal shows roughly the expected field dependence for a 2D noninteracting magnon gas; for strongly interacting magnon-phonon systems, a much stronger field dependence is expected,¹³ such as exhibited by the C_1CuCl sample. This indicates that, probably due to the larger number of dislocations, the

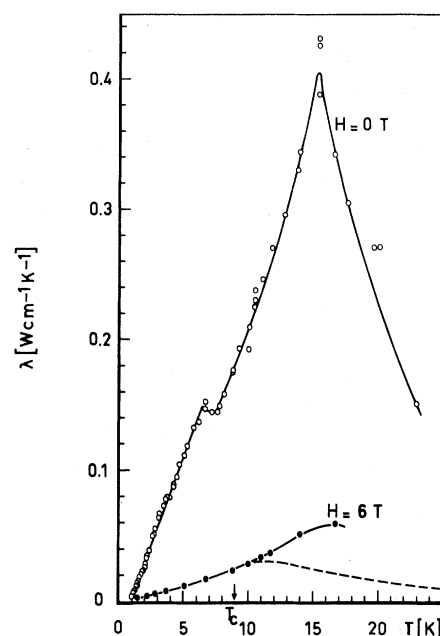


FIG. 3. Temperature dependence of the thermal conductivity of C_1CuCl at $H=0$ and 6 T. The broken line represents the thermal conductivity of the diamagnetic C_1CdCl , properly scaled to fit the 6-T curve below T_c .

C_1CuCl crystal has a much stronger magnon-phonon interaction than the C_2CuCl sample. This again implies that the absence of magnetic conduction above T_c in the C_2CuCl crystal is not due to a small intrinsic magnetic conductivity, but to a lack of contact between the magnetic system and the lattice.¹⁴

We would like to emphasize that some aspects of our experimental results could be explained with a model based on interactions between a phonon system and a magnetic system. However, taking into account the results of other very extensive studies on these compounds,^{4,7,9} which are consistent with our interpretation, we conclude that scattering between phonons and magnetic systems play a minor role in our experiments.

In conclusion, we have shown experimentally that in 2D Heisenberg ferromagnets, the magnetic system can contribute considerably to the energy transport, even at temperatures far above T_c . The magnitude of this contribution, as observed by thermal-conductivity measurements, is strongly dependent on the strength of the magnon-phonon interaction of the crystals.

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Superfluid-Vortex Thresholds and the Vortex-Core Parameter near T_λ

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Superfluid-vortex thresholds in a rotating annulus were detected with second sound in the temperature range $0.01 \text{ K} < T_\lambda - T < 0.2 \text{ K}$. A rapid decrease of the threshold angular velocity ω_{c1} was observed as $T \rightarrow T_\lambda$. The data can only be fitted to the theory of Fetter if a rapidly diverging vortex-core parameter is assumed.

Numerous investigators¹⁻³ have shown that experimental values of ω_{c1} , the threshold angular velocity for equilibrium vorticity in a rotating container of liquid helium, are in good agreement with the predictions of Fetter.⁴ In addition, the growth of vorticity above ω_{c1} in a rotating annulus has been shown⁵ to be in good agreement with the theory of Stauffer and Fetter.⁶ All of these investigations were made at temperatures between 1.2 and 1.8 K. In this Letter we report measurements of ω_{c1} in a rotating annulus at temperatures near the superfluid phase transition ($0.01 \text{ K} < \Delta T < 0.2 \text{ K}$; $\Delta T = T_\lambda - T$). The results are of interest because a rapid decrease of ω_{c1} with increasing temperature has been observed. The data can be fitted by Fetter's formula only if a rapidly diverging core parameter is assumed, which attains values near T_λ which are appreciably larger than generally assumed.

Fetter predicted, using a thermodynamic argument, that for a thin annulus of width D and mean radius R , the threshold is given by

$$\omega_{c1} = \frac{\kappa}{\pi D^2} \left(\ln \frac{2D}{\pi a} \right) \left(1 - \frac{D}{6R} \right), \quad (1)$$

where $\kappa = h/m_{\text{He}}$ is the quantum of circulation and a is the vortex-core parameter.⁷ The core parameter is generally assumed to be on the order of 1 \AA well below T_λ . Vortex-ring experiments have been used to determine a at low temperatures.^{8,9} Between 0.3 and 0.6 K (at saturated vapor pressure) a increases from 1.25 \AA to 1.35 \AA ; the total increase upon pressurization to the melting line is $\sim 30\%$. At higher temperatures, there are no precise measurements. It is often assumed that a is proportional to the "healing length" near T_λ , and hence that it diverges as $t^{-2/3}$, where $t = \Delta T/T_\lambda$.¹⁰ A search for this di-