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THE THREE DIMENSIONAL MAGNETIC STRUCTURE OF $CsNiF_3$: COMPARISON OF THE MEASURED SHORT RANGE ORDER WITH DIFFERENT MODELS

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The neutron-diffraction below 4K from a $CsNiF_3$-single crystal shows a Néel-point at 2.65K. The three dimensional order and the temperature dependence of the sublattice magnetization is discussed.

In the second part the already published experimental data for the susceptibility[1,2] and the correlation length are compared with different models. The discrepancy between the experimental and the theoretical data is briefly discussed.

1. NEUTRON DIFFRACTION FROM SINGLE CRYSTAL AT $T < 4.2$K

AT $T = 1.74$K the following magnetic Bragg-peaks were found (indexed in the chemical cell):

$$(1/2,0,0), \quad (3/2,0,0)$$
$$(-1/2,1,0)$$

The $(-1/2,1/2,0)$-reflection is absent. The $(1/2,0,0)$-reflection was found every 60° rotating the single crystal around the *c*-axis. This fact shows the existence of domains according to the hexagonal *c*-axis. From these magnetic reflections one can deduce the magnetic order; its projection onto the *a*-*b*-plane is shown in Fig. 1. The ferromagnetic order in the *c*-direction still exists. Therefore the three dimensional magnetic order is built up by ferromagnetic planes, which are coupled antiferromagnetically. This order is typical for systems with strong dipolar forces. The magnetization also shows that dipolar forces are important in the three dimensionally ordered $CsNiF_3$.[3]

Figure 2 shows the temperature-dependence of the integrated intensity for the $(1/2,0,0)$-reflection. These measurements show a Néel-point at

$$T_N = 2.67\text{K} \pm 0.05\text{K}$$

on a relative scale. The absolute error may be about ± 0.1K. A recent specific heat measurement done by Lebesque[4] shows a Néel-point at

$$T_N = 2.61\text{K} \pm 0.01\text{K},$$

which is in good agreement with the neutron diffraction result.

Fitting the neutron diffraction data to a power law of the form

$$I/I_0 = (T_N - T)^{2\beta}/T_N$$

one finds for $1.74\text{K} \leqslant T \leqslant 2.45\text{K}$

$$\beta = 0.3$$

and for $2.45\text{K} < T < 2.64\text{K}$

$$\beta = 0.5.$$

From the low temperature region a value for $I_0 = I(T = 0\text{K})$ is found, which corresponds to a magnetic moment per Ni^{2+}-ion of $2.25\mu_B$. This value is in good agreement with the saturation - moment per ion found from the magnetization measurements of $2.2\mu_B$.[2]

The deviation of β from the expected value 1/3 in the temperature-range $2.45\text{K} < T \leqslant 2.64\text{K}$ perhaps depends on the highly developed

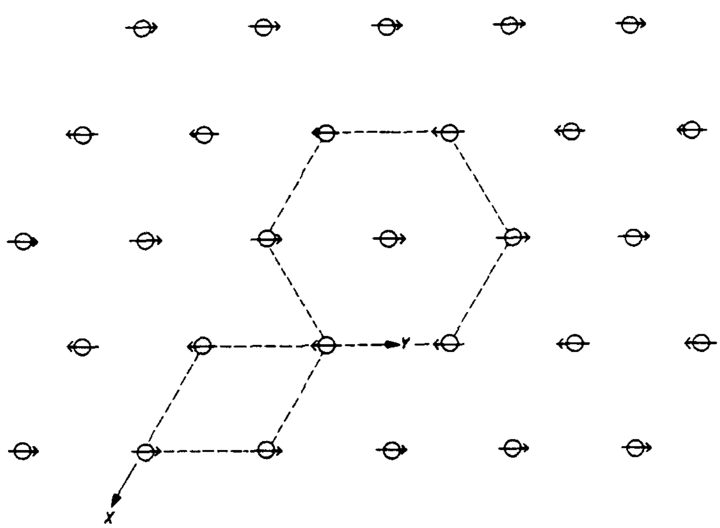

FIG. 1. Three dimensional magnetic order projected onto the *a*-*b*-plane.

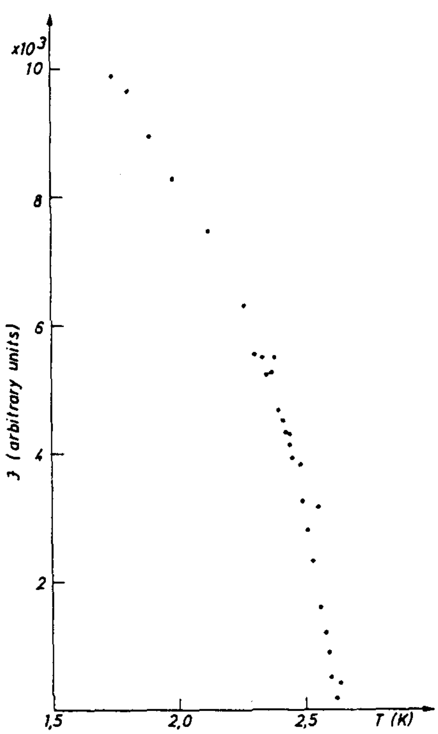

FIG. 2. Temperature-dependence of the integrated intensity for the (1/2,0,0) magnetic Bragg-peak.

ferromagnetic short range order, which leads to strong dipolar forces. But the determination of β must be done with more accuracy by new measurements to discuss this problem in more detail.

2. COMPARISON OF THE SUSCEPTIBILITY AND NEUTRON DIFFRACTION DATA FOR $T > 5$K WITH DIFFERENT THEORETICAL MODELS FOR THE FERROMAGNETIC LINEAR CHAIN

1. The susceptibility data are compared with the results for the MFA, the classical Heisenberg model, the classical planar Heisenberg model and the Ising model for $S = 1$.

(a) MFA

The MFA-results are identical with the high-temperature expansions of the linear models.

Using the formulas given by Domb and Miedema[5] the experimental data for $T \geqslant 100$K give an effective moment of

$$\mu_{eff} = 3.6$$

and an averaged exchange interaction I/k_{MFA} of

$$I/k_{MFA} = 3.5\text{K}$$

I/k_{MFA} should be the interaction in the chain, if the interchain-interaction is small compared with the intrachain-interaction.

(b) *Ising model with $S = 1$*

Using the formula of Obokata,[6] Oguchi or Suzuki *et al.*[7] the experimental data for

$7K \leqslant T \leqslant 40K$ give

$$I/k_{I,S} = 2.5K.$$

(c) *Classical Heisenberg model*

Fisher's formula[8] gives with the experimental data for $7K \leqslant T \leqslant 40K$

$$I/k_{H,S} = 6.5K.$$

(d) *Classical planar Heisenberg model*

Using Wegner's formula for the pair correlation[9] one can calculate the susceptibility. A fit for I/k gives

$$I/k_{pH,S} = 7K.$$

All results for the intrachain-interaction evaluated from the susceptibility-measurements are shown in Table 1.

Table 1. I/k evaluated from the susceptibility with different models

Model	I/k [K]
MFA	3.5
Ising, $S = 1$	2.5
Class. Heisenberg	6.5
Class. planar Heisenberg	7.0

2. From the neutron-diffraction data measured in the one-dimensional temperature-range $4K < T \leqslant 15K$ the correlation length in the ferromagnetic chains were evaluated.[10] These correlation lengths are compared with the Ising model, the classical Heisenberg model and the classical planar Heisenberg model using the pair-correlation function.

(a) *Ising model with* $S = 1$

Using the formula of Lieb, Schultz and Mattis[11] with $S = 1$ the experimental data give

$$I/k_{I,C} = 4.1K.$$

(b) *Classical Heisenberg model*

Fisher's formula for the pair-correlation function[8] gives

$$I/k_{H,C} = 19.5K.$$

(c) *Classical planar Heisenberg model*

Wegner's formula for the pair-correlation function[9] gives

$$I/k_{pH,C} = 18.1K.$$

In Table 2 all results for I/k are shown together.

Table 2. Values for I/k for different models evaluated from different experimental methods

	Model	I/k [K]
Susceptibility	MFA	3.5
	Ising	2.5
	Heisenberg	6.5
	Planar Heisenberg	7.0
Correlation length	Ising	4.1
	Heisenberg	19.5
	Planar Heisenberg	18.1

DISCUSSION

The first point, which should be emphasized, is: the MFA and the Ising model for $S = 1$ give nearly consistent values for I/k from the susceptibility and from the correlation length, whereas the Heisenberg model gives very divergent values for I/k from the susceptibility and the correlation length. The second point is: the results of the fits show[3] that in both cases the Heisenberg model describes the functional dependence over a great temperature-range quite well, whereas the Ising model fits the experimental data only in a small temperature-range.

Although the Ising model gives consistent values for I/k, one cannot say that the Ising model describes the one dimensional order in $CsNiF_3$, because the anisotropy measured in the magnetization[2] is typical for the planar Heisenberg model. Therefore the discrepancies in the values presented above for the intrachain interaction indicate that other effects like dipolar forces and perhaps a three dimensional short range order are important. New and more accurate measurement of the short range order and new theoretical calculations are necessary to explain the ferromagnetic short range order in $CsNiF_3$.

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REFERENCES

1. STEINER M., KRUGER W. and BABEL D., *Solid State Commun.* **9**, 327 (1971).
2. STEINER M., *Z. angew. Phys.* **32**, 116 (1971).
3. A detailed discussion will be published elsewhere.
4. LEBESQUE J.V., Private communication.
5. DOMB C. and MIEDEMA A.R., *Prog. Low Temperature Physics* Vol. IV, p. 296 (edited by GORTER) North Holland, Amsterdam (1964).
6. OBOKATA T. and OGUCHI I., *J. Phys. Soc. Japan* **25**, 322 (1968).
7. SUZUKI M., TSUYJAMA B. and KATSURA S., *J. Math. Phys.* **8**, 124 (1967).
8. FISHER M.E., *Am. J. Phys.* **32**, 343 (1964).
9. WEGNER R., *Z. Phys.* **206**, 465 (1967).
10. STEINER M. and DACHS H., *Solid State Commun.* **9**, 1603 (1971).
11. LIEB E., SCHULTZ T. and MATTIS D., *Ann. Phys.* **16**, 407 (1961).

Es wird über neue Ergebnisse der Neutronenbeugung bei Temperaturen unter 4K berichtet. Diese Messungen ergeben einen Néel-Punkt bei 2,65K. Die dreidimensionale Ordnung wird diskutiert.

Im zweiten Teil werden früher veröffentlichte experimentelle Ergebnisse für die Suszeptibilität und Korrelationslänge mit theoretischen Aussagen für verschiedene Modelle verglichen. Der Unterschied zwischen theoretischen und experimentellen Aussagen wird diskutiert.