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Neutron Scattering Studies of Low Dimensional
Magnetic Systems

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Within the last few years, neutron scattering studies have been carried out on several new prototypes of low dimensional (2D or 1D) magnetic systems. We will discuss in some detail a 2D Ising antiferromagnet K_2CoF_4 (Ikeda and Hirakawa), as well as our current results on K_2NiF_4 , $Rb_{2Mn_{0.5}Ni_{0.5}F_4}$ and $Rb_{2Mn_{0.5}Mg_{0.5}F_4}$. We will also report our new results of high resolution studies of TMMC. Recent neutron experiments at Brookhaven have been carried out in collaboration with J. Als-Nielsen (Risø), R. A. Cowley (Edinburgh), and H. J. Guggenheim (Bell Laboratories).

I. INTRODUCTION

Since the last Intern. Magnetism Conf. in Moscow 1973, we have witnessed significant progress in the neutron scattering studies of low dimensional magnetic systems. In two-dimensional (2D) magnetism, Ikeda and Hirakawa¹ reported their remarkable results on the 2D Ising antiferromagnet K_2CoF_4 ; / this experiment shows complete agreement with the exact solutions of Onsager.² Previously, a Heisenberg antiferromagnet K_2NiF_4 had been thoroughly investigated and its 2D characteristics were demonstrated in its static as well as dynamic magnetic properties.³ It appears now that "pure" 2D systems are reasonably well understood. Very recently, a systematic investigation was initiated for random 2D antiferromagnets such as $Rb_2Mn_{0.5}Ni_{0.5}F_4$ and $Rb_2Mn_{0.5}Mg_{0.5}F_4$.

In one-dimensional (1D) magnetic systems, definitive experiments were carried out on two model systems. Endoh et al⁴ studied the spin dynamics in $CuCl_2 \cdot 2NC_5D_5$, a 1D $S = \frac{1}{2}$ Heisenberg antiferromagnet. Well-defined excitations are observed over the whole zone with energies in agreement with the des Cloizeaux-Pearson exact solution. The second 1D example is $CsNiF_3$ investigated by Steiner et al.⁵ This represents an easy plane ferromagnetic chain and it shows different responses in "in-plane" and "out-of-plane" correlations.

In this brief review, we will discuss these neutron scattering results in some detail. Two comprehensive and excellent review articles have recently treated some of these model systems. In 1974, de Jong and Miedema⁶ gave detailed accounts of both 1 and 2D systems. The K_2CoF_4 study appeared after this review. On the other hand, both

$\text{CuCl}_2 \cdot 2\text{NC}_5\text{D}_5$ and CsNiF_3 were covered extensively in a very recent review article on 1D systems by Steiner et al.⁷ Thus we will not discuss these two compounds. Finally, we will present some new results of high resolution studies of $(\text{CD}_3)_4\text{N MnCl}_3$ (TMMC), a 1D $S = \frac{5}{2}$ Heisenberg antiferromagnet.⁸ These reveal interesting features in the spin dynamics at longer wavelengths than previously investigated.

II. 2D ISING SYSTEM K_2CoF_4

Neutron scattering studies on 2D magnetic systems started out, in 1969, with a near Heisenberg antiferromagnetic K_2NiF_4 .³ Then they were extended to Rb_2MnF_4 ³ and K_2MnF_4 .⁴ These measurements demonstrated unambiguously almost ideal 2D characteristics in all aspects of scattering function $\mathcal{S}(\mathbf{Q}, \omega)$. Observed critical quantities β, γ, ν could not be compared directly with Onsager's exact solutions since the latter apply to the Ising system. It was then highly desirable to find a compound which is planar and Ising-like in its interaction. As it turned out, K_2CoF_4 and Rb_2CoF_4 exhibit these characteristics.

The spin Hamiltonian for these compounds may be written

$$\mathcal{H} = \sum_{i>j} J_{\parallel} S_i^z S_j^z + J_{\perp} (S_i^x S_j^x + S_i^y S_j^y)$$

with fictitious spin $S = \frac{1}{2}$. Breed et al.¹⁰ have shown that $J_{\parallel}/J_{\perp} \sim 4$ for these 2D Co compounds. However, it was neutron experiment itself which demonstrated these compounds to be almost ideal Ising systems. Samuelsen¹¹ reported neutron scattering measurements on Rb_2CoF_4 and, independently, Hirakawa and his collaborator has carried out extensive studies on K_2CoF_4 not only by neutron technique but also by specific

heat measurement.¹²

Fig. 1 shows the sublattice magnetization M_s of K_2CoF_4 measured by Ikeda and Hirakawa. It is truly remarkable that, for the entire temperature range, M_s follows Onsager's analytic formula

$$\frac{M_s(T)}{M_o} = \left(1 - \sinh^{-4} \frac{2J}{kT}\right)^{1/8}$$

Near $T_N = 108$ K, the magnetization can be approximated by

$$\frac{M_s(T)}{M_o} = B \left(\frac{T_N - T}{T_N}\right)^\beta$$

with $\beta = 0.123 \pm 0.008$, in excellent agreement with the exact solution of 0.125. It is quite instructive to compare this result with those for Heisenberg antiferromagnets K_2NiF_4 and K_2MnF_4 , which are also shown schematically in Fig. 1 (see also Table I). The exponent β are quite similar; 0.14 for K_2NiF_4 ³ and 0.15 for K_2MnF_4 .¹³ However, low temperature behavior of M_s is entirely different, reflecting the difference between Heisenberg and Ising systems.

The other critical exponents are also in good agreement with the theoretical prediction. The cross sections above T_N were analyzed by the formula of Fisher and Burford¹⁴

$$\chi(q) = \frac{A(q)}{(\kappa ||^2 + q^2)^{1-\frac{\eta}{2}}}$$

where $A(q)$ is a slowly changing function with temperature. Here, $T_N, \kappa ||$ and $\chi(0)$ may be expressed as

$$\kappa || \propto \left(\frac{T - T_N}{T_N} \right)^\nu, \quad \chi(0) \propto \left(\frac{T - T_N}{T_N} \right)^\gamma$$

They obtained $\nu = 0.97$; $\gamma = 1.71$ and $\eta = 0.2$ in excellent agreement with Onsager's exact solutions (see Table I). In addition, the specific heat of K_2CoF_4 and Rb_2CoF_4 was investigated by Ikeda et al.¹² Again they observed a symmetric logarithmic singularity as predicted by the theory.

III. 2D HEISENBERG ANTIFERROMAGNET

In the previous subsection we have discussed the experiments on the highly anisotropic 2D antiferromagnets K_2CoF_4 and Rb_2CoF_4 . As we have seen, there is good absolute agreement with the exact results of Onsager for the 2D Ising model. These experiments employed techniques developed earlier in an extensive study of the 2D near-Heisenberg antiferromagnet K_2NiF_4 by Birgeneau, Skalyo, and Shirane.¹⁵ Somewhat ironically, it has turned out that the analysis of the 2D Heisenberg systems is much more complex, and it is only recently that the critical behavior of these systems has been properly elucidated. In particular, the apparent mean field-like exponents $\gamma \sim 1$, $\nu \sim \frac{1}{2}$ originally determined by Birgeneau et al.¹⁵ for K_2NiF_4 are incorrect due to an improper treatment of the

transverse susceptibility. In this subsection, we discuss recent experiments and analysis by Als-Nielsen, Birgeneau, Guggenheim, and Shirane¹⁶ on K_2NiF_4 , K_2MnF_4 and $Rb_2Mn_{0.5}Ni_{0.5}F_4$.

The spin Hamiltonian for these systems may be written

$$\mathcal{H} = \sum_{i>j} J_{ij} S_i S_j + \sum_i g_i \mu_B H_i^A S_i^Z$$

The exchange is predominantly between nearest neighbors alone. The anisotropy field H_i^A originates from a combination of dipolar spin-spin interactions and single-ion crystal-field anisotropy. The strength of the Ising field relative to the isotropic exchange field varies from 0.2 to 0.7% for the three compounds.

The general behavior of the fluctuations in these systems have been extensively reviewed previously¹⁵, so we shall not repeat that here. We simply remind the reader that above T_N the crystal exhibits a ridge of critical scattering where the ridge is uniform in the direction perpendicular to the sheets. In the critical region, only the zz component of the wave vector dependent susceptibility $\chi^{\alpha\alpha}(\vec{q})$ is divergent. At and below T_N a 3D Bragg peak appears, but the critical scattering remains 2D. As noted in the previous section, in K_2NiF_4 and K_2MnF_4 the sublattice magnetization follows the simple power law

with $\beta = 0.138 \pm 0.004$ and 0.15 ± 0.01 , respectively. In both cases $B \sim 1.0$. The random system is complicated by some smearing of T_N . Empirically, it is found that the amount of smearing scales approximately with the size of the crystal. In their smallest sample Als-Nielsen et al found that the Bragg intensity is accurately described by the square of the above power law convoluted with a Gaussian distribution of Néel temperatures. Least squares fits yield $\beta = 0.16 \pm 0.01$, $B = 1.0$, $\langle T_N \rangle = 68.72 \pm 0.02$ K and a relative Gaussian smearing of $\sigma/\langle T_N \rangle = 0.6\%$. It is evident that both B and β are extremely close, if not identical, to those in the pure systems. Furthermore, β is close to the asymptotic 2D Ising value of 0.125.

We now consider the critical scattering measurements in the three systems for $T > T_N$; these were performed in the $(h, 0, \ell)$ plane, where ℓ is the coordinate perpendicular to the sheets. For purely two-dimensional spin correlations the critical scattering should take the form of ridges along ℓ at $h = 1, 3, \dots$. At all temperatures, aside from known geometrical effects, the critical scattering is indeed independent of ℓ . For each system the detailed measurements across the ridge were carried out around $(1, 0, \frac{1}{2})$, where, for an incident neutron energy of 13.6 meV, the quasielastic approximation becomes nearly exact. The cross section at this position is given by

$$\sigma(q) = T | 0.95 \chi^{\parallel}(q) + 1.05 \chi^{\perp}(q) |$$

Here $\parallel$ and $\perp$ are with respect to the Ising axis and $q = (q_x^2 + q_y^2)^{1/2}$.

We now consider the appropriate functional form for $\chi(q)$. Tracy and McCoy¹⁷ have shown that for the 2D Ising model the Lorentzian approximation $\chi(q) = A(t)/(q^2 + \kappa^2)$ is essentially exact for $q \leq 10 \kappa$. In order to keep the analysis of the results at the simplest level, Als-Nielsen et al discard data with $q > 10$ and assume a cross section $\sigma(q)$ of the form

$$\sigma(q) = T | A^{\parallel} / (q^2 + \kappa^{\parallel 2}) + A^{\perp} / (q^2 + \kappa^{\perp 2}) |$$

The most complex aspect of the data analysis originates in the difficulty of separating out $\chi^{\parallel}$ and $\chi^{\perp}$. In their original study Birgeneau et al assumed that $\chi^{\perp}$ could be absorbed into the background near T_N ; this assumption was later proved to be unjustified. As discussed in detail in recent papers we believe that the above technique remedied this difficulty with $\kappa^{\perp}$. The results for the correlation length $\kappa^{\parallel}$ are shown in Fig. 2 together with the corresponding data for the pure materials K_2NiF_4 and K_2MnF_4 . From the figures it is evident that over the reduced temperature range studied, the critical behavior is identical in all three systems. Detailed fits to simple power laws for the correlation length and staggered susceptibility yield $\nu = 0.9 \pm 0.1$ and $\gamma = 1.65 \pm 0.15$, respectively, for all three systems. We do not regard the differences from the ideal 2D Ising values of 1.0 and 1.75 as

significant given both the approximations in the data analysis and the possible presence of higher order corrections to the asymptotic power laws.

In summary, then, the near-Heisenberg 2D antiferromagnets of the K_2NiF_4 class all exhibit 2D Ising-like phase transitions driven by the Ising anisotropy, whether single-ion or dipolar in origin. Randomness appears to have no measurable effect on the observed critical behavior.

On the other hand, the dynamic properties¹⁰ exhibit rather striking effects for $Rb_2Mn_{0.5}Ni_{0.5}F_4$ due to its random nature of the system.

Fig. 3 shows the results of inelastic measurements for $Rb_2Mn_{0.5}Ni_{0.5}F_4$. They are characterized by two well-defined bands of excitations. The width and energies of the zone boundary modes are accurately predicted using a simple Ising cluster model while overall dispersion is correctly given by the Walker mean-crystal model (see Ref.18). An intensive algebraic and "computer experiment" analysis has been carried out by Kirkpatrick and Harris¹⁹ in good overall agreement with the experiment.

IV. HIGH RESOLUTION STUDY OF TMMC

In the last five years extensive work has been done on the dynamics of one-dimensional systems. Experiments have been concentrated on three model systems:

- (1) $(\text{CD}_3)_4\text{NMnCl}_3$ (TMMC), a 1D $S = 5/2$ Heisenberg antiferromagnet
- (2) $\text{CuCl}_2 \cdot 2\text{NC}_5\text{D}_5$, a 1D $S = 1/2$ Heisenberg antiferromagnet
- (3) CsNiF_3 , a 1D easy plane $S = 1$ Heisenberg ferromagnet

Experiments and theory appropriate to these three systems have recently been reviewed in detail by Steiner, Villain, and Windsor.⁷ We shall therefore limit our discussion in the section to a report of some new high resolution measurements on TMMC.

The exchange Hamiltonian for TMMC may be written⁸

$$\mathcal{H} = - 2J_{\text{nn}} \sum_i \vec{S}_i \cdot \vec{S}_{i+1}$$

with $J_{\text{nn}} = 6.5$ K. In the classical spin approximation²⁰ the static inverse correlation length is given simply by

$$a\kappa_s = \frac{1 + u}{(-u)^{1/2}}$$

$$\text{where } u = \coth K - 1/K, K = \frac{2J_{\text{nn}} S(S+1)}{k_B T}$$

where a is the nearest neighbor Mn^{++} spin separation.

In their original measurements Hutchings et al⁸ showed that at $T = 1.9 \text{ K}$ ($T_N = 0.845 \text{ K}$) TMMC exhibits well-defined spin-wave excitations throughout most of the second half of the 1D Brillouin zone that is for $\frac{1}{2} \frac{\pi}{a} \leq q \leq 0.95 \frac{\pi}{a}$. Further, the q -dependence of the intensities is well described by simple spin-wave theory. At 1.9 K the inverse correlation length is approximately $\kappa_5 = 0.005 \frac{\pi}{a}$ so that the existence of well-defined propagating modes in the above q -range need not be considered surprising.

For completeness we have extended the measurements of Hutchings et al to cover the range

$$0.1 \frac{\pi}{a} \leq q \leq 0.985 \frac{\pi}{a}$$

at $T = 1.45 \text{ K}$. Again sharp excitations are observed over this whole range. The consequent dispersion relation is shown in Fig. 4. As expected a double-sine curve is obtained with the periodicity of the antiferromagnetic Brillouin zone, that is, with a double periodicity in the true nuclear Brillouin zone appropriate to the paramagnetic phase. Once more, the excitation intensities are accurately predicted by simple classical spin-wave theory.

Hutchings et al also reported some measurements of the temperature dependence of the spin excitations between 1.9 K and 40 K at $q = 0.95 \frac{\pi}{a}$ and $q = 0.9 \frac{\pi}{a}$. At both wave vectors the excitation becomes progressively broader as the temperature is increased ultimately becoming overdamped at the higher temperatures. As discussed by Steiner et al⁷, these measurements stimulated an enormous amount of theoretical activity

on 1D paramagnetic spin dynamics. By now an impressive array of theories have appeared and, not surprisingly, all are relatively successful in accounting for the above-mentioned results. However, since the Hutchings et al experiments are at rather large wave vectors, they do not really provide a very critical test of the theories. In order to resolve a number of basic physical questions as well as to provide a substantive test of current and future theories, we have performed high resolution measurements at low temperatures and long wavelengths in TMMC. We have chosen the scans so as to probe two basic questions:

(1) Is there any evidence for a strong dynamic central peak arising from "correlated spin cluster" diffusion?

(2) Is there a dynamic critical wave vector κ_0 characterizing the evolution from underdamped to overdamped behavior as a function of q and how is it related to the static inverse correlation length κ_s .

Measurements at $q^* = \frac{\pi}{a} - q = 0.015, 0.03, \text{ and } 0.04 \frac{\pi}{a}$ were performed using 5.5 meV incident neutrons. Background, including the incoherent scattering around $E = 0$, was determined by several measurements near $q \sim \frac{\pi}{2a}$ where no low energy magnetic scattering is expected. This background was subtracted point-by-point from the small q^* data. The experimental results after this subtraction are shown for $q^* = 0.015, 0.03 \frac{\pi}{a}$ in Figs. 5,6. These data have not been corrected for any instrumental factors. In order to transform the data to the form $\mathcal{S}(\vec{q}, \omega)$ for detailed comparison with theory the data should be multiplied by the factor $(\frac{5.5}{5.5 - \hbar\omega})^2$. The instrumental resolution is 0.13 meV, full width at half maximum (FWHM). Measurements were also performed at $q^* = 0.05 \frac{\pi}{a}$ using 13.7 meV incident neutrons. A similar background subtraction was carried out but in this case the incoherent scattering around $E = 0$

was too large to allow meaningful results to be obtained near $E = 0$. These data have been corrected for instrumental intensity factors so that they represent the true Van Hove scattering function $\mathcal{L}(\vec{q}, \omega)$ convoluted with an instrumental resolution function with FWHM 0.60 meV. The $q^* = 0.05 \frac{\pi}{a}$ results are shown in Fig. 7.

It is evident that these experiments provide direct answers to both questions (1) and (2). Firstly, somewhat to our surprise, there is no evidence for a distinct central peak at $q^* = 0.015 \frac{\pi}{a}$ or $q^* = 0.03 \frac{\pi}{a}$ at any temperature. This would appear to place severe restrictions on theories based on "pseudo-static" clusters. This absence of a central peak appears to be unique to one-dimension since in both two- and three-dimensional Heisenberg systems three peaked functions are generally observed above T_N . Secondly, at each wave vector it is apparent that as q^* goes from $\sim 2 \kappa_s$ to κ_s the dynamic line shape changes from being peaked at finite energy to being overdamped. Thus there is indeed a dynamic critical wave vector K_c and, not surprisingly, it is simply related to the static inverse correlation length κ_s . From Figs. 5-7, one would estimate $\kappa_c \sim 1.5 \kappa_s$.

This contradicts explicitly the Lovesey-Meserve theory²¹, which predicts κ_c varies with square root of κ_s . Correspondingly the Lovesey-Meserve theory predicts diffusive behavior through the q, T range covered by Figs. 5 and 6 in disagreement with the experiment.

There are also a number of theories²² which assume $\kappa_c = \kappa_s$ as a starting point and further have built into them the assumption that the system will exhibit propagating excitations for $q > \kappa_s$. These are all somewhat unsatisfactory in that they assume much of the empirical answer. Nevertheless, they do provide an important first step towards a genuine first-principles theory. Such calculation has been performed by McLean and Blume²² for $q^* = 0.015 \frac{\pi}{a}$. The agreement is only partially satisfactory. It would undoubtedly be improved considerably by choosing $\kappa_c \simeq 1.5 \kappa_s$. Clearly, much further work remains to be done on 1D spin dynamics. We hope that these new results will provide valuable intuitive guidance to future theories as well as a substantive testing ground for quantitative comparison.

V. CONCLUDING REMARKS

We have discussed several recent studies of low dimensional magnetic systems. We have now representative prototypes of 1D and 2D model systems well characterized by neutron scattering techniques. Recently, attention has been directed towards the percolation transition in magnetic model systems. Here again low dimensional systems may play a vital role because of their simplicity. Experimental work is now underway for the $\text{Rb}_2\text{Mn}_c\text{Mg}_{1-c}\text{F}_4$ system.²³

For the square lattice with nn interactions alone, the percolation limit is $C_p = 0.59$. The experiments have been carried out on two specimens with $C = 0.54 \pm 0.02$ and 0.57 ± 0.02 . The critical scattering displays fascinating characteristics and a preliminary report will be given by Cowley et al.²³ in this conference. We limit ourselves to a brief mention of dynamical properties. The spectrum of $C = 0.54$ sample shows well-defined peaks / as shown in Fig. 8. They are also quite different from those observed in $\text{Rb}_2\text{Mn}_{0.5}\text{Ni}_{0.5}\text{F}_4$ (Fig. 3). The observed cross sections are excellently described by computer simulation by Alben and Thorpe²⁴ based on the linear spin-wave approximation. Further experiments along these lines are now in progress.

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Table I

Critical exponents for 2D magnetic systems

	β	ν	γ	Reference
2D Ising	0.125	1.0	1.75	Onsager (2)
K_2CoF_4	0.123	0.97	1.71	Ikeda & Hirakawa (1)
K_2NiF_4	0.14	0.9	1.65	Als-Nielsen et al (16)
$Rb_2Mn_{0.5}Ni_{0.5}F_4$	0.16	0.9	1.65	"

FIGURE CAPTIONS

- Fig. 1 Sublattice magnetization for $K_0\text{CoF}_4$, after Ikeda and Hirakawa.⁽¹⁾ The solid line marked as 2D Ising is the Onsager's exact solution. Observed curves for other antiferromagnets are shown schematically.
- Fig. 2 Inverse correlation lengths for three 2D antiferromagnets, together with the exact result for 2D Ising model, after Als-Nielsen et al.⁽¹⁶⁾
- Fig. 3 Dispersion relations in the $[\pi 00]$ direction for $\text{Rb}_2\text{Mn}_{0.5}\text{Ni}_{0.5}\text{F}_4$, together with those for two end members. After Als-Nielsen.⁽¹⁶⁾
- Fig. 4 Entire dispersion curve for TMMC spin waves at $T = 1.45\text{K}$.
- Fig. 5 High resolution data for TMMC at $q^* = 0.015 \frac{\pi}{a}$
- Fig. 6 High resolution data for TMMC at $q^* = 0.03 \frac{\pi}{a}$
- Fig. 7 Neutron energy gain and loss spectrum for TMMC at $q^* = 0.05 \frac{\pi}{a}$
- Fig. 8 Cross sections for $\text{Rb}_2\text{Mn}_{0.54}\text{Mg}_{0.46}\text{F}_4$. Solid lines are computer simulation by Alben and Thorpe²⁴ for this composition.

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5

FIGURE 6

FIGURE 7

NEUTRON SCATTERING FROM $\text{Rb}_2\text{Mn}_{0.54}\text{Mg}_{0.46}\text{F}_4$ AT 4.0 K AS
COMPARED WITH COMPUTER SIMULATIONS

FIGURE 8