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Abstract

With the discovery of magnetic ordering in $RbMnF_3$, this unique antiferromagnetic system was recognized as a prime case for a test of conventional spin-wave theory (CSWT) because of its negligibly small anisotropy and its simple, cubic structure. CSWT predicts a simple T^2 power-law fall-off of the sublattice magnetization. Yet to this day, no stringent tests have been made of this prediction. Seiden [(Phys. Lett. **28 A**, 239 (1968)] deduced a T^3 low-temperature behavior on the basis of antiferromagnetic resonance measurements, concluding that CSWT was not supported. We have recently carried out neutron scattering measurements of both single-crystal and powdered samples of $RbMnF_3$ in order to test for CSWT, Seiden's result, and two other more recent semiempirical spin-wave schemes, and we present an analysis of the results. Measurements in the critical regime gave values of the critical exponent β and of T_N that are in agreement with previous measurements.

Disciplines

Physical Sciences and Mathematics | Physics

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Neutron scattering measurements in $RbMnF_3$: A test of spin-wave-region theories at low temperatures and critical behavior near T_N

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With the discovery of magnetic ordering in $RbMnF_3$, this unique antiferromagnetic system was recognized as a prime case for a test of conventional spin-wave theory (CSWT) because of its negligibly small anisotropy and its simple, cubic structure. CSWT predicts a simple T^2 power-law fall-off of the sublattice magnetization. Yet to this day, no stringent tests have been made of this prediction. Seiden [(Phys. Lett. **28 A**, 239 (1968)] deduced a T^3 low-temperature behavior on the basis of antiferromagnetic resonance measurements, concluding that CSWT was not supported. We have recently carried out neutron scattering measurements of both single-crystal and powdered samples of $RbMnF_3$ in order to test for CSWT, Seiden's result, and two other more recent semiempirical spin-wave schemes, and we present an analysis of the results. Measurements in the critical regime gave values of the critical exponent β and of T_N that are in agreement with previous measurements. © *2012 American Institute of Physics*. [doi:10.1063/1.3679410]

After the discovery of antiferromagnetism[1] in $RbMnF_3$, it was realized that $RbMnF_3$ constituted quite a unique realization of a near-ideal cubic antiferromagnet, because of the smallness of the energy gap between the ground state and its magnetic excitation spectrum, and that as a result was an excellent candidate for making a stringent experimental test of the predictions of conventional spin-wave theory (CSWT). CSWT applied to antiferromagnets predicts a T^2 power-law fall-off of the sublattice magnetization (SLM) from its $T = 0$ value. If the energy gap of an antiferromagnet is not small, as is the case for nearly all antiferromagnets found so far, the "gap effects" predicted by CSWT completely wash out a power-law fall-off and preclude the experimental detection of any definitive power-law fall-off. In $RbMnF_3$ the gap effects are essentially absent, and in this sense $RbMnF_3$ is unique.

One of the original investigators of the energy gap in $RbMnF_3$, Seiden,[2] tried to test the low-temperature power-law fall-off prediction of CSWT for $RbMnF_3$ by doing antiferromagnetic resonance (AFMR) measurements and concluded that the SLM fell off at T^3, instead of the predicted T^2. Clearly, a T^3 fall-off constitutes solid evidence against the validity of CSWT, and that was what Seiden concluded. He bolstered his case by citing[2] the ^{55}Mn NMR measurements of Teaney, which purportedly also showed a T^3 dependence. Because Seiden's analysis of the AFMR data was based on certain possibly dubious assumptions, however, and because Teaney's results were never published, no significance was attributed to their conclusions thereafter.

Recently, Koebler and collaborators (see Ref. 3) and, separately, Bykovetz *et al.*[4] have advanced the view that essentially all magnetic systems empirically appear to show simple power-law fall-offs of the magnetization (or SLM), with few caveats. The semiempirical schemes proposed by Koebler,[3] and alternatively by Bykovetz,[4] to explain the observed power laws differ in both the empirical determinations of the claimed power-law exponents and the proposed explanations for these power-law behaviors. Bykovetz identifies separate and distinct power-laws[4,5] for ferromagnetic and antiferromagnetic systems in the "low-temperature" region $M_0 < M < 0.90M_0$ (and retains a spin-wave perspective), whereas Koebler claims a more general power-law universality[3] for all magnetic systems, with power-law behaviors persisting in some cases as far as $\sim$0.85 T_c (Koebler's proposed power laws depend on the spin of the magnetic ions and the dimensionality and isotropy properties of the magnetic systems).

Because $RbMnF_3$ is such a unique case, in that it is expected from every one of the above perspectives to exhibit a simple power law behavior in the "low temperature" region, we decided to re-visit the case of $RbMnF_3$ experimentally in order to see which, if any, of the four predictions/observations would be supported by experiment.

Specifically, Koebler's scheme predicts that an isotropic magnetic system, with a half-integral spin, should be characterized by the same simple T^2 fall-off of the SLM as in CSWT, except that the fall-off should persist much closer to T_c (as far as $\sim$0.8 T_c).[3] By contrast, in Bykovetz's scheme a simple antiferromagnet like $RbMnF_3$ should exhibit either a $T^{2.29}$, $T^{2.66}$, or T^4 fall-off[5] in the magnetization region $M_0 < M < 0.90M_0$ (which for $RbMnF_3$ translates to temperatures below $\sim$0.5 T_N). The properties that determine which of the three exponents prevails in a given system have not yet been identified, except *a posteriori*.

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To be sure, the ideal way to make precise determinations of power-law exponents would be to do NMR measurements, because NMR allows extremely high precision. However, in the case of $RbMnF_3$, nature has conspired to make the situation very difficult. The transferred hyperfine fields at the locations of both the Rb and the F ions cancel out in the $RbMnF_3$ magnetic structure. But, although the ^{55}Mn NMR does exist,[6] it is plagued by enormous "frequency-pulling" effects (precisely *because of* the smallness of the energy gap in $RbMnF_3$), and so no one, apart from Teaney (see Ref. 2), has even tried to attempt to measure the NMR temperature dependence as yet.

As a consequence, we decided to carry out neutron scattering (NS) measurements, as that is the next best method for establishing whether or not $RbMnF_3$ does exhibit a power-law behavior. Initial measurements were made on a large single crystal of $RbMnF_3$, and subsequent ones on a powdered sample produced by crushing a small piece of the same crystal. Because of the high perfection of the single crystal (mosaic spread of 0.08°), extinction effects were unfortunately too large to correct for. Two separate runs of Bragg-reflection data were then obtained for the crushed-crystal sample, a lower-temperature run (1.5 to ~70 K) and a critical region run (70 to 88 K). The first-run data are presented in numerical form in Table I. Various power-law fits were then made using the power-law equation $M = M_0 + BT^C$ and assuming $M \propto \sqrt{I}$.

If one fits the entire data set of Table I to a single power law (i.e., up to $T \sim 0.85\ T_c$), one obtains $M = 147.6 + 0.00131\ T^{2.47 \pm 0.02}$. Figure 1 shows a plot of the data versus $T^{2.47}$, showing an excellent fit. Thus, the power-law fit for this entire temperature range appears to give clear-cut evidence for an exponent of 2.5. This appears at first glance to rule out Koebler's scheme. However, Koebler *et al.*[7] observed the exact same result in NS done on MnF_2. An exponent of 2.5 is one of the powers expected in Koebler's scheme,[3] but for the case of anisotropic, half-integral-spin magnetic systems. Thus, unless it can be shown that there is significant distortion of the cubic structure of $RbMnF_3$ leading to anisotropic behavior, our result constitutes evidence against the validity of Koebler's scheme. Koebler has proposed doing synchrotron measurements[8] in order to find evidence of distortion of the cubic $RbMnF_3$ structure. Such a distortion, however, must be shown to be greater than that in EuO, EuS, and EuTe, because Koebler has argued that these three cubic compounds are isotropic and all purportedly display a T^2 behavior. It should be noted that evidence acquired in earlier research investigations indicates no evidence for anisotropic behavior or distortion of the cubic structure in $RbMnF_3$. In particular, previous measurements show virtually no magnetostriction.[9] Likewise, ^{55}Mn NMR showed no detectable trace of quadrupole splitting,[6] indicating that a perfectly cubic structure exists at 4.2 K.

TABLE I. Neutron scattering intensities vs temperature (K).

T	I	T	I	T	I
1.51	21 961.2	26.29	20 496.1	49.19	16 313.1
1.59	21 794.4	27.29	20 385.5	50.18	16 081.5
5.12	21 727.4	28.28	20 413.9	51.18	15 765.0
6.11	21 997.1	29.28	20 144.6	52.17	15 525.1
7.12	21 674.8	30.28	19 984.3	53.17	15 212.5
8.11	21 676.6	31.29	19 862.5	54.16	15 045.4
9.14	21 454.1	32.28	19 714.7	55.16	14 787.9
10.15	21 529.1	33.30	19 568.9	56.15	14 401.1
11.17	21 517.1	34.27	19 332.3	57.15	14 282.4
12.18	21 625.1	35.28	19 198.1	58.15	13 942.2
13.19	21 510.6	36.27	19 176.2	59.15	13 527.2
14.19	21 534.6	37.27	18 786.1	60.15	13 281.3
15.20	21 533.9	38.26	18 783.3	61.14	12 981.9
16.21	21 435.0	39.26	18 502.8	62.14	12 539.9
17.22	21 273.2	40.24	18 337.2	63.14	12 353.4
18.23	21 315.6	41.26	18 141.3	64.16	11 925.7
19.26	21 369.0	42.24	17 878.2	65.15	11 658.7
20.26	21 132.2	43.24	17 631.3	66.15	11 282.0
21.28	20 938.5	44.22	17 291.9	67.14	10 924.6
22.28	21 096.7	45.22	17 271.5	68.70	10 370.1
23.30	20 934.1	46.21	17 094.9	69.79	10 061.6
24.28	20 791.7	47.20	16 716.2	71.03	9526.9
25.30	20 734.9	48.19	16 502.4		

In neutron measurements[7] on MnF_2, Koebler *et al.* reported a $T^{2.5}$ fall-off, which was attributed to MnF_2's being an anisotropic half-integral spin system. However, careful examination of precision ^{19}F NMR measurements[10] showed that in the region of $20\,K \le T \le 43\,K$, the magnetization curve of MnF_2 deviates from the $T^{2.5}$ behavior, giving instead an exponent of 2.29 ± 0.02 (in accord with Bykovetz's scheme). Additionally, below ~20 K, the magnetization curve fits, over a significant region, a power law with an exponent of 2.67 ± 0.02. Thus, because the NS measurements have much less precision than NMR, these relatively small deviations in the exponent from the value of 2.5 do not make themselves visible within the scatter of the NS data points.

It is worthwhile to mention at this point that in conventional analyses of the magnetization curve, the "intermediate region," i.e., the region between the spin-wave region (usually the ~10% fall-off below saturation) and the critical region, is not well characterized theoretically. Because of the current work, we were led to observe that empirically it appears that an "intermediate region" exists in seemingly all

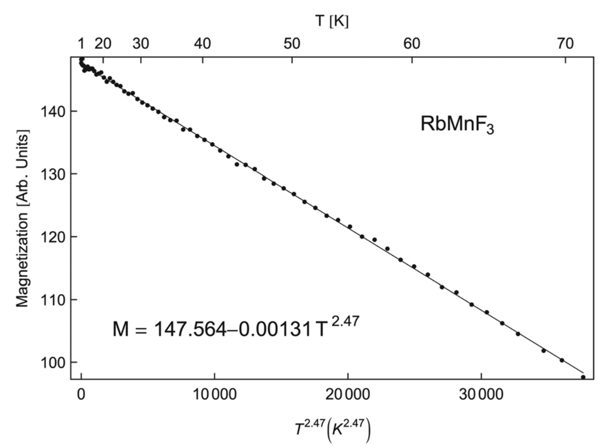

FIG. 1. $RbMnF_3$ sublattice magnetization fall-off fitted to a single power-law for the entire Table I data set, with M taken as $\propto \sqrt{I}$.

cases (where precision NMR data are available) characterized by a $T^{2.5}$ power-law fall-off in the temperature range of roughly $T > 0.5\ T_c$ to $0.75\ T_c$, as is the case in EuO,[11] EuS,[12] and MnF_2.[10] Yet in these cases, the magnetization curve *below* $\sim 0.5\ T_c$ exhibits discernibly different power-laws. Thus, when the data are not sufficiently precise, the entire curve below $\sim 0.75\ T_c$ might give the appearance of following a single $T^{2.5}$ power-law fall-off while masking the lower-temperature deviations within the scatter.

We see, therefore, that even a simple attempt to characterize a curve with power-laws is fraught with pitfalls. Specifically, unless the functional (e.g., power-law) behavior is known to be valid *a priori*, the least-squares fitting process, by itself, can become very misleading by showing an apparently superb fit (with small errors) for cases in which a single power-law dependence does not actually exist. The above-mentioned fits of the MnF_2 data, which appear to give a superb fit to $T^{2.5}$ for the NS data,[7] are a case in point, whereas the more precise NMR data show a change in functional dependence in different temperature ranges.[10]

With the above in mind, we tested our data versus CSWT, as well as Seiden's observations, looking at the temperature range below ~25 K, where CSWT should certainly apply (cf. Seiden[2]). Unfortunately, the scatter in our data is too large to give a precise power-law exponent, or even to verify that a simple power-law prevails here. Our data do, however, appear to definitely exclude the purported AFMR and NMR observation of a T^3 behavior reported in Ref. 2. Graphical analysis (i.e., the plotting of the data for various different exponents until a straight line is obtained) shows that within the scatter of the data, the T^2 behavior expected according to CSWT is not inconsistent with our data for $T \leq 25$ K (cf. Seiden's measurements[2]). However, although a least-squares fit to the data for $T \leq 25$ K does give an exponent of ≈ 2, the uncertainty is quite large (± 0.4).

Lastly, to find out whether one of the power-laws from the scheme of Bykovetz[5] might be appropriate to describe the region $M_0 < M < 0.90 M_0$ (which translates into the temperature range of $T < \sim 0.6\ T_N$), we omitted the data below 22 K (because of gross scatter) and fitted the smooth part of the data curve ($22\ K \leq T \leq 50\ K$). The least-squares fit to this data gives an exponent of 2.26 ± 0.09, which is clearly consistent with an exponent of 2.29, which was also the behavior observed in the NMR data of MnF_2.[10] Fig. 2 shows our data plotted versus $T^{2.29}$, showing a good fit to all points below ~50 K.

Our second run, carried out in the critical region (70 to 88 K), was done in order to check the critical behavior. A fit of the data to the critical region equation $M/M_o = D(1 - T/T_N)^{\beta}$ gave an exponent $\beta = 0.33 \pm 0.02$ and $T_N = 82.6$ K. Both values agree with previous measurements. A measurement to determine β was also made with the single crystal but gave a value of 0.16, presumably because of the observed severe extinction effects.

In summary, we have carried out detailed NS measurements on a crushed crystal sample and found the following results. The T^3 power-law behavior deduced by Seiden[2] from AFMR measurements is ruled out by our data. The T^2 behavior expected in Koebler's scheme[3] is contradicted by our results unless, contrary to previous measurements, it can be

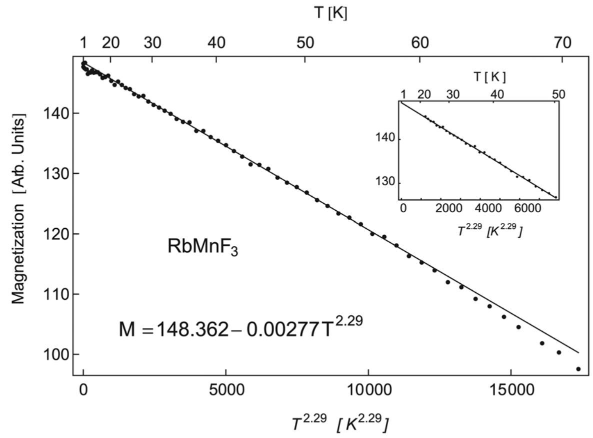

FIG. 2. $RbMnF_3$ sublattice magnetization plotted as a function of $T^{2.29}$. Inset shows the $22\ K < T < 50\ K$ data region used in the fit.

shown that $RbMnF_3$ is anisotropic. The data below 25 K are *not* incompatible with the T^2 behavior predicted by CSWT, but our NS measurements do not allow any convincing validation, either. Our data are compatible with Bykovetz's scheme if the power-law is $T^{2.29}$ (a behavior previously observed in the NMR of antiferromagnetic MnF_2, as well as of EuTe), but more precise data are needed in order for definitive conclusions to be made.

We firmly believe that the accumulation of good magnetization data (mostly NMR) in many magnetic systems[3,4] over a number of decades points to the existence of low-temperature simple power-law behaviors, at least in simple magnetic systems, contrary to the expectations of CSWT. Although $RbMnF_3$ still remains the prime antiferromagnetic candidate for a good test of the various schemes (most especially CSWT), higher precision (<0.1%) than in our NS measurements (~0.5%) is required. In view of the above NS results, it would be highly desirable to carry out the difficult ^{55}Mn NMR measurements so that the uniqueness of $RbMnF_3$ could be exploited fully, at least in terms of making a long-overdue stringent test of CSWT.

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