

## Dipolar anisotropy in quadratic-layer antiferromagnets

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Dipolar interactions in quadratic-layer Heisenberg antiferromagnets have been treated in the spin-wave approximation to calculate the temperature dependence of the sublattice magnetization and the magnon energy gap. Magnon-magnon interactions are included to first order. Numerical results for  $K_2MnF_4$  and the double-layer  $K_3Mn_2F_7$  confirm the earlier spin-wave analyses of these structures, in which the  $k=0$  magnon energies were made dependent on temperature in a semi-empirical way. The gaps as a function of the temperature scale with the sublattice magnetizations.

### I. INTRODUCTION

The principal quantities entering the spin-wave analysis of Heisenberg antiferromagnets, including the two-dimensional (2D) ones, are the nearest-neighbor exchange and the anisotropy. The anisotropy, even when weak, has a profound effect on the magnon dispersion near the center of the Brillouin zone, and thus on thermodynamic quantities, by invoking a gap at  $k=0$ . The gap decreases with temperature, and ultimately drops to 0 at the transition to the paramagnetic state. In particular in the case of 2D systems, antiferromagnetic resonance<sup>1,2</sup> (AFMR) as well as neutron scattering<sup>3</sup> have shown the  $k=0$  energy to closely follow the sublattice magnetization. This also holds for the Mn compounds, in which the anisotropy is mainly of dipolar origin. In a spin-wave theory, however, dipolar interactions are difficult to treat because they involve many pairs of spins. The usual approach therefore is to represent the anisotropy by a staggered magnetic field,<sup>4</sup> despite its failure to account for anisotropy-induced interactions among the spin waves. With increasing tem-

perature such interactions act to lower the spin-wave energies near  $k=0$ , as observed, whereas a staggered field leaves the  $k=0$  gap nearly constant. In order to imitate the temperature dependence of the gap, spin-wave renormalization near  $k=0$  is incorporated in a *semiempirical* way by allowing the staggered field itself to fall with temperature. In the remaining part of the Brillouin-zone renormalizing corrections according to Oguchi,<sup>5</sup> primarily due to the exchange, are predominant. The staggered-field approach has provided an adequate spin-wave description at low temperatures.<sup>6,7</sup>

The primary purpose of this paper is to show that in a 2D spin-wave theory that includes dipolar anisotropy the  $k \approx 0$  modes are properly renormalized without making the spin-Hamiltonian parameters dependent on temperature. In a spin-wave expansion, the four-magnon part of the anisotropy, i.e., the part to lowest order responsible for the temperature dependence of the gap, is treated as a perturbation. In the actual calculations, the systems considered are the single-layered  $K_2MnF_4$  ( $T_N=42.1$  K) and the double-layered  $K_3Mn_2F_7$  ( $T_N=58.3$  K).

### II. SPIN WAVES IN SINGLE-LAYER STRUCTURES

The calculation of the magnon dispersion in quadratic layers, and the sublattice magnetization derived from it, is based on a model of nearest-neighbor antiferromagnetic Heisenberg exchange and a weak anisotropy by dipolar interactions summed over the lattice. For completeness, we further include axial single-ion anisotropy<sup>8</sup> and an external field along the easy axis. The Hamiltonian thus reads

$$\mathcal{H} = |J| \sum_{l,m} \vec{S}_l \cdot \vec{S}_m + \frac{1}{2} g^2 \mu_B^2 \sum_{\substack{i,j \\ i \neq j}} [\vec{S}_i \cdot \vec{S}_j / r_{ij}^3 - 3(\vec{S}_i \cdot \vec{r}_{ij})(\vec{S}_j \cdot \vec{r}_{ij}) / r_{ij}^5] + D_s \sum_i (S_i^z)^2 - g\mu_B H_0 \sum_i S_i^z, \quad (1)$$

where the indices  $l$  and  $m$  refer to neighboring sites on the up and down sublattices, respectively;  $i$  and  $j$  run over both sublattices.

Following the standard procedure, we expand the spin operators in local Holstein-Primakoff spin-deviation operators  $a_l$  and  $b_m$ , and subsequently go over to reciprocal space. The quadratic part of the Hamiltonian may then be written as  $\mathcal{H}_0 + \mathcal{H}_1$ , with<sup>9-11</sup>

$$\mathcal{H}_0 = \sum_{\vec{k}} [(T + g\mu_B H_0) a_{\vec{k}}^\dagger a_{\vec{k}} + (T - g\mu_B H_0) b_{\vec{k}}^\dagger b_{\vec{k}} + G(a_{\vec{k}}^\dagger b_{\vec{k}} + a_{\vec{k}} b_{\vec{k}}^\dagger)],$$

$$T = 4|J|S + A(\vec{k}) + C - 2D_s S, \quad (2)$$

$$G = 4|J|S\gamma_{\vec{k}} + D(\vec{k}),$$

$$\mathcal{H}_1 = \sum_{\vec{k}} [B(\vec{k})(a_{\vec{k}} a_{-\vec{k}} + b_{\vec{k}}^\dagger b_{-\vec{k}}^\dagger) + E(\vec{k})a_{\vec{k}} b_{-\vec{k}}^\dagger + \text{c.c.}] .$$

For the quadratic layer  $\gamma_{\vec{k}} = \cos(\frac{1}{2}k_x a)\cos(\frac{1}{2}k_y a)$ , with  $a$  the dimension of the 2D magnetic cell. Equations (2) contain the lattice summations, in the notation of Harris,<sup>10</sup>

$$\begin{aligned} A(\vec{k}) &= d \sum_n (3 \cos^2 \theta_n - 1) (1 + \frac{1}{2} e^{i\vec{k} \cdot \vec{R}_n}) / R_n^3, \\ B(\vec{k}) &= -\frac{3}{4} d \sum_n \sin^2 \theta_n e^{-2i\phi_n} e^{i\vec{k} \cdot \vec{R}_n} / R_n^3, \\ C &= -d \sum_n' (3 \cos^2 \theta_n - 1) / R_n^3, \\ D(\vec{k}) &= \frac{1}{2} d \sum_n' (3 \cos^2 \theta_n - 1) e^{i\vec{k} \cdot \vec{R}_n} / R_n^3, \\ E(\vec{k}) &= -\frac{3}{2} d \sum_n' \sin^2 \theta_n e^{-2i\phi_n} e^{i\vec{k} \cdot \vec{R}_n} / R_n^3, \end{aligned} \quad (3)$$

with  $d = g^2 \mu_B^2 S$ , and  $\sum_n$  and  $\sum_n'$  denoting summations over magnetic sites on the same and opposite sublattices, respectively. In Eqs. (2) and (3),  $\vec{k}$  is taken a three-dimensional (3D) vector to include dipolar interactions among the layers.

Because  $\mathcal{H}_1$  (comprising dipolar terms only) mixes  $\vec{k}$  and  $-\vec{k}$ ,  $\mathcal{H}_0 + \mathcal{H}_1$  would require  $4 \times 4$  Bogoliubov transformation for diagonalization.  $\mathcal{H}_0$  by itself, on the other hand, may be diagonalized by a  $2 \times 2$  transformation of the standard form

$$a_{\vec{k}} = u_{\vec{k}} \alpha_{\vec{k}} - v_{\vec{k}} \beta_{\vec{k}}^\dagger, \quad b_{\vec{k}} = -v_{\vec{k}} \alpha_{\vec{k}}^\dagger + u_{\vec{k}} \beta_{\vec{k}}. \quad (4)$$

The substantial complications introduced by  $\mathcal{H}_1$  when dealing with the  $1/2S$  corrections below may thus be avoided by treating  $\mathcal{H}_1$  as a perturbation to  $\mathcal{H}_0$ . Note that in  $\text{K}_2\text{MnF}_4$  the anisotropy, mainly of dipolar origin, is  $\sim 4 \times 10^{-3}$  in units  $4|J|S$ , or  $\sim 0.3$  K, compared to  $T=0$  magnon energies ranging from 7.4 to 84 K.<sup>7</sup> In zero field, then,  $\mathcal{H}_1$  lifts the degeneracy to first order by shifts amounting to approximately  $\pm |E - 2\gamma_{\vec{k}} B| / \epsilon_{\vec{k}}$ , in which  $\epsilon_{\vec{k}}$  is the magnon energy associated with  $\mathcal{H}_0$  in

units  $4|J|S$ . In  $\text{K}_2\text{MnF}_4$  the splitting vanishes at the zone center as both  $B$  and  $E$  are zero there, and increases to  $\pm 5$  parts in  $10^3$  at the zone boundary. Splittings of this size are of no concern when calculating spin-wave gaps and thermodynamic quantities. For  $\text{K}_2\text{MnF}_4$  in zero field this has been documented more precisely in Ref. 7, where the energies from exact diagonalization of  $\mathcal{H}_0 + \mathcal{H}_1$ ,<sup>9</sup>

$$\begin{aligned} E_{\vec{k}} = \{ & T^2 - G^2 + EE^* - 4BB^* + (g\mu_B H_0)^2 \\ & \pm 2[(T^2 - G^2)(g\mu_B H_0)^2 \\ & + (ET - 2BG)(E^*T - 2B^*G) \\ & + (B^*E - BE^*)^2]^{1/2} \}^{1/2}, \end{aligned} \quad (5)$$

have, in approximate form, been compared with those from  $\mathcal{H}_0$  alone. In second order the energies are shifted by  $\sim (EE^* - 4BB^*)/8|J|S\epsilon_{\vec{k}}$ , which again is 0 at  $k=0$ .  $\mathcal{H}_1$  will not be considered further, but we note that Eq. (5) may be used to include its effects to lowest order in  $1/2S$ . With reference to Eq. (1), ignoring  $\mathcal{H}_1$  amounts to neglect of the nonsecular terms of the dipolar interaction.

In order for the gap to fall with temperature, it is essential to consider magnon-magnon interactions. This is accomplished in the usual way by including the four-magnon part of the expansion of  $\mathcal{H}$  in  $1/2S$  as a first-order perturbation.<sup>5</sup> However, to simplify the algebra we apply the random-phase approximation (RPA) prior to diagonalization to obtain an effective two-magnon Hamiltonian.<sup>12</sup> The transformation diagonalizing the latter has the form of the one diagonalizing  $\mathcal{H}_0$ , Eq. (4), but modified coefficients. The results are, to first order in  $1/2S$ , equivalent to those of the standard Oguchi scheme. Retaining only those terms which ultimately contribute to the magnon dispersion, we arrive at the effective two-magnon Hamiltonian

$$\begin{aligned} \mathcal{H} = \sum_{\vec{k}} [ & Z_1(\vec{k}) a_{\vec{k}}^\dagger a_{\vec{k}} + Z_2(\vec{k}) b_{\vec{k}}^\dagger b_{\vec{k}} \\ & + Z_3(\vec{k}) (a_{\vec{k}} b_{\vec{k}} + a_{\vec{k}}^\dagger b_{\vec{k}}^\dagger) ], \end{aligned} \quad (6)$$

in which

$$\begin{aligned} Z_1(\vec{k}) &= 4|J|S + A(\vec{k}) + C - (2S - 1)D_s + g\mu_B H_0 \\ & - \frac{1}{NS} \sum_{\vec{k}'} \{ (4|J|S + C)\mu_2(\vec{k}') + [4|J|S\gamma_{\vec{k}} + D(\vec{k})]\mu_3(\vec{k}') + [A(\vec{k}) + 4SD_s]\mu_1(\vec{k}') \\ & + [A(\vec{k}') + 2A(\vec{k} - \vec{k}') - 2A(\vec{0})][\mu_1(\vec{k}') + \frac{1}{2}] \}, \\ Z_3(\vec{k}) &= 4|J|S\gamma_{\vec{k}} + D(\vec{k}) - \frac{1}{NS} \sum_{\vec{k}'} \{ \frac{1}{2}[4|J|S\gamma_{\vec{k}} + D(\vec{k})][\mu_1(\vec{k}') + \mu_2(\vec{k}')] \\ & + [4|J|S\gamma_{\vec{k}}\gamma_{\vec{k}'} - 2D(\vec{k} - \vec{k}')] \mu_3(\vec{k}') \}, \end{aligned} \quad (7)$$

while  $Z_2(\vec{k})$  is obtained from  $Z_1(\vec{k})$  by inverting the sign in front of  $g\mu_B H_0$  and interchanging  $\mu_1(\vec{k})$  and  $\mu_2(\vec{k})$ . In Eqs. (7),  $\mu_1(\vec{k}) = \langle a_{\vec{k}}^\dagger a_{\vec{k}} \rangle$ ,  $\mu_2(\vec{k}) = \langle b_{\vec{k}}^\dagger b_{\vec{k}} \rangle$ , and  $\mu_3(\vec{k}) = \langle a_{\vec{k}} b_{\vec{k}} \rangle = \langle a_{\vec{k}}^\dagger b_{\vec{k}}^\dagger \rangle$ ;  $N$  is the number of magnetic unit cells. The Hamiltonian Eq. (6) is diagonalized by Eq. (4) with

$$u_{\vec{k}} = [\frac{1}{2}(F_{\vec{k}} + 1)]^{1/2}, \quad v_{\vec{k}} = [\frac{1}{2}(F_{\vec{k}} - 1)]^{1/2}. \quad (8)$$

Here,

$$F_{\vec{k}} = [Z_1(\vec{k}) + Z_2(\vec{k})] / 2\Omega_{\vec{k}}, \quad (9)$$

$$\Omega_{\vec{k}} = \{ \frac{1}{4} [Z_1(\vec{k}) + Z_2(\vec{k})]^2 - [Z_3(\vec{k})]^2 \}^{1/2}. \quad (10)$$

The magnon energies then become

$$E_{\vec{k}}^{(1,2)} = \Omega_{\vec{k}} \pm \frac{1}{2} [Z_1(\vec{k}) - Z_2(\vec{k})]. \quad (11)$$

Inserting the Bogoliubov transformation Eq. (4) with the above  $u_{\vec{k}}$  and  $v_{\vec{k}}$ , we further have

$$\begin{aligned} \mu_1(\vec{k}) &= \frac{1}{2} F_{\vec{k}} (n_{\vec{k}}^{(1)} + n_{\vec{k}}^{(2)} + 1) + \frac{1}{2} (n_{\vec{k}}^{(1)} - n_{\vec{k}}^{(2)} - 1), \\ \mu_2(\vec{k}) &= \frac{1}{2} F_{\vec{k}} (n_{\vec{k}}^{(1)} + n_{\vec{k}}^{(2)} + 1) + \frac{1}{2} (n_{\vec{k}}^{(2)} - n_{\vec{k}}^{(1)} - 1), \\ \mu_3(\vec{k}) &= -\frac{1}{2} G_{\vec{k}} (n_{\vec{k}}^{(1)} + n_{\vec{k}}^{(2)} + 1), \end{aligned} \quad (12)$$

where  $G_{\vec{k}} = Z_3(\vec{k}) / \Omega_{\vec{k}}$ , and  $n_{\vec{k}}^{(1,2)} = 1 / [\exp(E_{\vec{k}}^{(1,2)} / k_B T) - 1]$  are the Bose occupation numbers of the magnon branches.

The magnetizations of the up and down sublattices are finally given by

$$\begin{aligned} \langle S_l^z \rangle &= S - \Delta_0 - \Delta S_0(T) + \Delta S_1(H_0, T), \\ \langle S_m^z \rangle &= -S + \Delta_0 + \Delta S_0(T) + \Delta S_1(H_0, T), \end{aligned} \quad (13)$$

respectively, in which

$$\Delta S_0(T) = \frac{1}{2N} \sum_{\vec{k}} F_{\vec{k}} (n_{\vec{k}}^{(1)} + n_{\vec{k}}^{(2)}), \quad (14)$$

$$\Delta S_1(H_0, T) = \frac{1}{2N} \sum_{\vec{k}} (n_{\vec{k}}^{(1)} - n_{\vec{k}}^{(2)}), \quad (15)$$

while the zero-point spin reduction reads

$$\Delta_0 = \frac{1}{2N} \sum_{\vec{k}} (F_{\vec{k}} - 1). \quad (16)$$

### III. COMPARISON WITH EXPERIMENTS IN $K_2MnF_4$

In this section we discuss the computer evaluation of the magnon energies and the sublattice magnetization in  $K_2MnF_4$  ( $S = \frac{5}{2}$ ,  $g \approx g_e$ ), and compare the results with experimental data and the earlier analysis based on a semiempirical anisotropy field. As for the parameters, we take  $|J|/k_B = 8.41$  K, which is the weighted average from various experiments in both the ordered and paramagnetic regime.<sup>7,13-15</sup> The magnetic lattice param-

eters at 4.2 K are  $a = (4.151 \pm 0.003)\sqrt{2}$  Å and  $c = 13.242 \pm 0.010$  Å.<sup>16</sup>

Our first concern is to evaluate the dipolar summations, Eqs. (3). Except for  $C$ , which is independent of  $\vec{k}$ , the summations were determined at a selection of  $100 \times 100$  equidistant points in the 2D Brillouin zone by summing over the lattice within a Lorentz sphere of radius  $20a$ . Advantage was taken of the symmetry, and the convergence was checked by summing out to  $30a$  in a number of cases. For arbitrary  $\vec{k}$ , then, the dipolar sums were found by linear interpolation. Two specific points should be commented on. First, in case  $k \approx 0$  the summations do not converge to a unique value when extended beyond the Lorentz sphere.<sup>10,17</sup> Here, we notice that the  $k=0$  energy depends, to leading order in  $1/2S$ , on the summations in the combination  $A + C - D$ , contributions to which from the continuum outside the Lorentz sphere are essentially 0. A similar conclusion holds with regard to the  $1/2S$  corrections. Second, and more important, the summations depend on  $k_z$ . We have examined in some detail the question of how dipolar interactions among the layers affect, as a function of  $k_z$ , the dispersion. This would be very cumbersome with inclusion of the  $1/2S$  corrections, but it is expected that a determination of the energy at ( $k_x=0, k_y=0$ ) on the basis of  $\mathcal{H}_0$ , i.e., the leading terms of  $Z_i(\vec{k})$ , will provide a reliable assessment of the effects. When varying  $k_z$  from the center of the zone out to the zone boundary, corresponding to going from in-phase to out-of-phase precession of adjoining layers, it appears that the 2D gap does increase, but by 4 parts in  $10^5$  only. We henceforth set  $k_z=0$ .

Given the temperature, the external field, and the parameters of the system, the coefficients  $Z_i(\vec{k})$ , and from them the energies and Bose occupation numbers, can now be evaluated as a function of  $\vec{k}$ . The calculation is done self-consistently since these quantities occur implicitly at the right-hand side of Eqs. (7). Typically, five iterations were required for sufficient convergence. Summations in Eqs. (6) of the form  $\sum_{\vec{k}'} A(\vec{k} - \vec{k}') \mu_1(\vec{k}')$ , which contain a shifted argument, were computed for  $50 \times 50$  points in the 2D Brillouin zone, and at other points obtained by interpolation. These summations are convolutions, and may therefore be efficiently performed via passage back to  $\vec{r}$  space. From the calculations it emerged that dipolar anisotropy cannot fully account for the experimental magnon gap. The residual anisotropy has been attributed to single-ion anisotropy upon noting that isolated  $Mn^{2+}$  in the nonmagnetic isomorphs of  $K_2MnF_4$  also exhibit a weak  $D_s$  term.<sup>18</sup> To determine  $D_s$ , the calculated zero-temperature gap was made coincident with the gap measured by AFMR,  $E_{k=0}(T=0)/k_B = 7.40 \pm 0.05$  K.<sup>2</sup> The result is  $D_s/k_B = (-9 \pm 2) \times 10^{-3}$  K, which is comparable with  $D_s$  of  $Mn^{2+}$  in the nonmagnetic hosts.

In Fig. 1 the sublattice magnetization  $\langle S^z \rangle$  is plotted, as calculated from Eq. (13) with the above parameters inserted. Near zero temperature we find  $\langle S^z \rangle = 2.329$ , or a zero-point spin reduction  $\Delta_0 = 0.171$ , to be compared with the experimental result  $\Delta_0 = 0.17 \pm 0.03$ ,<sup>19</sup> and  $\Delta_0 = 0.170$  from spin-wave theory with a staggered anisotropy field.<sup>7</sup> Also plotted in Fig. 1 are the data from NMR,<sup>7</sup> anchored

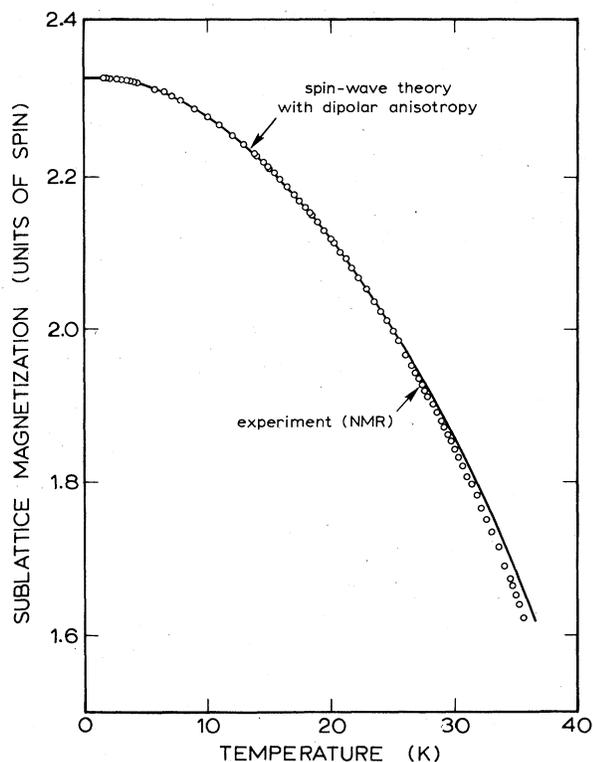


FIG. 1. Calculated sublattice magnetization versus temperature in  $K_2MnF_4$ , compared with the data from  $^{19}F$  NMR (Ref. 7).

to the calculated  $\langle S^z \rangle$  at  $T=0$  K. Excellent agreement is observed to exist up to about 20 K, or  $T/T_N \approx 0.5$ , which essentially is the upper limit of validity of the earlier spin-wave analysis based on a staggered field. In fact, the fall of  $\langle S^z \rangle$  with temperature calculated here corroborates the earlier analysis even above the point of departure from experiment. At 35 K, for instance,  $\langle S^z \rangle = 1.6886$ , compared to  $\langle S^z \rangle = 1.6878$  calculated in Ref. 7. Another salient result from the calculations is that the development of the spin-wave gap with temperature tracks the fall of the sublattice magnetization to great precision. Up to at least 35 K, the difference nowhere exceeds a few tenths of percent. In this way the relation  $E_{k=0}(T) \propto \langle S^z(T) \rangle$ , which is frequently adopted in spin-wave approaches based on a staggered field, is given a theoretical basis in case of dipolar anisotropy.

It is noteworthy that in the present analysis the gap at  $T=0$  is successfully taken to match the AFMR value. It has been pointed out, however,<sup>7</sup> that these quantities need not coincide because in a 2D spin-wave theory the gap is an effective one made up of the  $(k_x=0, k_y=0)$  energy averaged over all  $k_z$  rather than the energy at  $k=0$ . In the presence of a residual exchange along the  $c$  axis of magnitude  $J_c$ , the effective gap is larger by a factor  $1 + |J_c|S/H_A$ , where  $H_A$  represents the anisotropy. The  $T=0$  magnon gap here may indeed be varied slightly without significant detrimental effects on the fit, provided  $J$  is appropriately adjusted, and of course kept within its error bounds. This sets an upper limit to  $|J_c/J|$  of order  $10^{-4}$ .

To further explore the validity of the present analysis, we also calculated the magnetizations residing on the sublattices in an external field, and compared the results with both the data and the spin-wave analysis based on a staggered field of Ref. 15. Up to 20 kG and 20 K excellent agreement has been found for the individual sublattice magnetizations as measured with NMR. In this region, the present calculation also closely reproduces the spin-wave analysis of Ref. 15. With regard to the *net* magnetization in a field, our calculated results agree with the spin-wave analysis of Ref. 15 up to 30 kG.

#### IV. DOUBLE-LAYER STRUCTURES

The spin-wave theory of Sec. II may be expanded straightforwardly to Heisenberg double-layer antiferromagnets with dipolar anisotropy, of which  $K_3Mn_2F_7$  is the archetype.<sup>20</sup> The development of the theory is more complex because a primitive magnetic unit cell contains four spins, two up and two down. Accordingly, there are four sublattices. The magnon dispersion has two branches, an "acoustic" branch, corresponding to in-phase precession of the paired layers, and an "optical" branch, corresponding to out-of-phase precession and having a minimum energy  $\sim 4|J|S$ . The formalism will not be reproduced here. We note, however, that the Hamiltonian from which to start, the dipolar summations, and the final expressions for the energies, the sublattice magnetizations, and the zero-point spin reduction are appropriate generalizations of Eqs. (1), (3), (11), (13), and (16), respectively.

The results of the calculations for  $K_3Mn_2F_7$  (Fig. 2) as

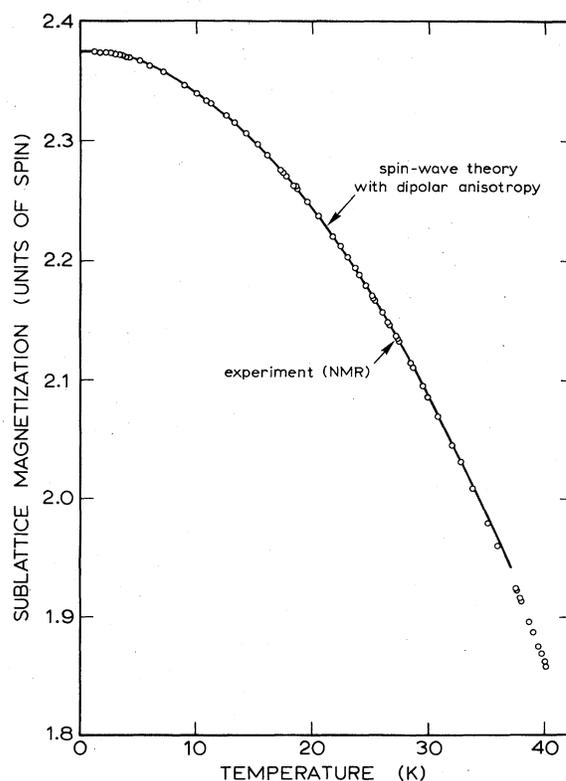


FIG. 2. Same as Fig. 1, but for the double-layer structure  $K_3Mn_2F_7$ . The NMR data are taken from Ref. 20.

well as the conclusions derived from them are very similar to those for the single-layer system  $K_2MnF_4$ . The calculations were done while setting the nearest-neighbor exchange parameter equal to  $|J|/k_B = 7.59$  K, as obtained from NMR.<sup>20</sup> The magnetic lattice parameters at 4.2 K are  $a = 4.181 \times \sqrt{2}$  Å and  $c = 21.55$  Å (Ref. 21); the separation between the paired layers was taken equal to the in-layer Mn-Mn spacing. We again made allowance for a small single-ion anisotropy such as to attain agreement of the calculated gap with the AFMR result at zero temperature. The latter is  $E_{k=0}(T=0)/k_B = 5.78 \pm 0.05$  K,<sup>22</sup> which yields  $D_s/k_B = (-6 \pm 2) \times 10^{-3}$  K. For the zero-point spin reduction we calculated  $\Delta_0 = 0.125$ , in excellent agreement with experiment,<sup>23</sup> and equal to the result of a spin-wave calculation based on a staggered anisotropy field. Accord with the semi-empirical staggered-field spin-wave formalism was also found for the temperature dependence of  $\langle S^z \rangle$ . As in Ref. 20, the departure of the present spin-wave analysis from the experimental data sets in at about 30 K (cf. Fig. 2). Also the calculated temperature dependence of the spin-wave gap is very similar to the case of  $K_2MnF_4$  in that it accurately follows the dependence of  $\langle S^z \rangle$ .

## V. CONCLUSIONS

We have calculated, as a function of the temperature, the magnon energies and the sublattice magnetizations in typical examples of quadratic single-layer and double-layer antiferromagnets with dipolar anisotropy. The spin-wave theory developed is based on Heisenberg exchange and dipolar interactions, and includes corrections to the magnon energies to first order in  $1/2S$ . In earlier work, where the anisotropy had been represented by a staggered field, spin-wave descriptions of ordered 2D systems were concluded to break down above  $\frac{1}{2}T_N$ . This conclusion is essentially confirmed. The region of the spin-wave fit has not been modified. An important distinction from the earlier treatment, however, is a faithful renormalization of the spin waves in the center of the Brillouin zone by the formalism itself rather than by allowing the gap to vary with temperature in a semiempirical way. This in fact leaves only the zero-temperature parameters to be adjusted in the spin-wave fit. The fall of the spin-wave gap with temperature has been found to follow the thermal reduction of the sublattice magnetization.

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