one might start with a Hamiltonian with an analytic cut off and preintegrate it over the region $1 < k < \infty$. We shall not go further into this problem here.

The final result for (A1) is thus

$$\text{const} - \frac{1}{4}\frac{1}{(a+b)^3}\frac{1}{(16\pi^2)^2} q^2 \ln 2 + O(q^4) + O(2-\sigma) + \text{terms}(q, q^3, \text{etc.}) \quad . \quad \text{(A5)}$$

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Spin-Wave Analysis of the Quadratic-Layer Antiferromagnets K_2NiF_4, K_2MnF_4, and Rb_2MnF_4

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In this and the following two papers, low-temperature spin-wave properties of quadratic-layer antiferromagnets having the K_2NiF_4 structure are reported and analyzed in detail. Here we present the results of a least-squares adjustment of spin-wave theory to the temperature variation of the sublattice magnetization in the compounds K_2NiF_4, K_2MnF_4, and Rb_2MnF_4, as reflected by ^{19}F NMR frequency measurements in zero field. Lowest-order temperature-dependent and temperature-independent corrections to simple spin-wave theory, as formulated by Oguchi, are included in the analysis. The free parameters of the fits are taken to be the exchange coupling, the zero-temperature spin-wave gap energy, and the zero-temperature ^{19}F NMR frequency. Our conclusions are as follows. Spin-wave theory accounts for the sublattice magnetization of these compounds up to somewhat less than one-half the Néel temperature, with the temperature-dependent corrections yielding less than 20% improvement in the range of fit for the Mn^{2+} compounds and a negligible improvement for K_2NiF_4. The breakdown of spin-wave theory is clearly not ascribable to spin-wave interaction effects and is apparently caused by excitations of a fundamentally different nature. Exchange values obtained are in excellent agreement with data from neutron and susceptibility measurements. The "effective" spin-wave-energy-gap values obtained give some evidence for interplanar exchange coupling between second-neighbor planes, yielding upper limits for such coupling of a few parts in 10^4 of the primary exchange. Earlier conclusions regarding the large zero-point spin reduction in K_2NiF_4 are refined here, giving a result slightly larger than but within error limits of the spin-wave-theory value (17.7%).

I. INTRODUCTION

The isomorphic compounds K_2NiF_4, K_2MnF_4 and Rb_2MnF_4, whose magnetic properties were extensively investigated by Breed and co-workers,[1] appear to be almost ideal two-dimensional (2D) antiferromagnets. The large separation between the planes, in which the magnetic ions form a quadratic lattice, and the symmetry relations between these planes of ions combine to make interplanar interactions between the magnetic ions extremely weak. In sharp contrast to a material such as $CrBr_3$, where the ratio of intraplanar to interplanar exchange is quite large, but of order 10, it is,

in the present materials, of the order of several thousand. The predominantly 2D character of the system was brought out most clearly in the neutron scattering experiments of Birgeneau *et al.*[2] Here it was shown that, as the temperature is lowered toward the transition temperature, there is increasing evidence for scattering by a short-range order which is entirely 2D in character. Only at the Néel temperature, where this 2D order becomes long range, does a 3D ordering of the planes become possible as a result of the dipolar and/or exchange interactions between them.

The predicted behavior of 2D spin arrays with *isotropic* Heisenberg interactions only has been considered by Mermin and Wagner[3] and by Stanley and Kaplan.[4] It was established rigorously by the former that such a system can have no long-range order at finite temperatures. There is no energy gap in the excitation spectrum and the situation at low temperatures is controlled by the density of states for small excitation energies or, in fact, for small $\vec{k}$ values. As the dimensionality of the system decreases, this low-$\vec{k}$ density increases and in two dimensions leads to a catastrophic excitation of spin waves which destroys the long-range order. In the presence of any anisotropy, such as to leave the system with an easy axis, the above argument is not valid. An energy gap appears in the excitation spectrum at $\vec{k}=\vec{0}$ and inhibits the disordering process at sufficiently low temperatures. Stanley and Kaplan[4] showed that an analysis of the high-temperature series for the susceptibility of the isotropic system predicted a divergence at a finite temperature, in the manner familiar in 3D Heisenberg systems or for the 2D Ising system. To reconcile their result with that of Mermin and Wagner, they suggested that the transition might be to a state without long-range order but having a divergent susceptibility. This is plausible from phase-space considerations. Consider, for simplicity, a ferromagnet whose average energy at some temperature is proportional to $-\sum_{\vec{k}}\gamma_{\vec{k}}\langle\vec{S}_{\vec{k}}\cdot\vec{S}_{-\vec{k}}\rangle$, where $\gamma_{\vec{k}}=(1/Z)\sum_{\vec{\delta}}e^{i\vec{k}\cdot\vec{\delta}}$, with $\vec{\delta}$ a nearest-neighbor displacement and $\vec{S}_{\vec{k}}$ the Fourier transform of the local spin vector $\vec{S}_i$. The quantity $\vec{S}_{\vec{k}}$ also has to satisfy the sum rule $\sum_{\vec{k}}\langle\vec{S}_{\vec{k}}\cdot\vec{S}_{-\vec{k}}\rangle=NS(S+1)$. As the temperature decreases, so does the average energy and since $\gamma_{\vec{k}}$ has its maximum at $\vec{k}=\vec{0}$, the system achieves this by increasing $\langle\vec{S}_{\vec{k}}\cdot\vec{S}_{-\vec{k}}\rangle$ for small $\vec{k}$. In a 3D system, there is such a paucity of low-$\vec{k}$ states that the system eventually is forced below a certain temperature into making $\langle\vec{S}_0\cdot\vec{S}_0\rangle$ of order N—that is to say it orders. A 2D system meets this situation by developing a pole of some order in $\langle\vec{S}_{\vec{k}}\cdot\vec{S}_{-\vec{k}}\rangle$ at $\vec{k}=0$, and this will imply infinite susceptibility, but no long-range order. Real systems possess anisotropy and thus may be expected to be ordered at sufficiently low temperatures. In the present cases the anisotropy is small (anisotropy field/exchange field $\sim 10^{-3}$) and the transition temperatures appear to be rather close to the Stanley–Kaplan values. It appears that the existence of the transition is controlled by the stronger exchange interactions and that the anisotropy then determines the nature of the new state below T_N.

The present paper and a number of others associated with it discuss various aspects of spin waves in the quadratic-layer (QL) compounds. These are the zero-point spin deviations,[5] the temperature dependence of sublattice magnetization (considered below),[6] antiferromagnetic resonance (AFMR) in high fields,[7] and the dispersion relation of the magnons as a function of temperature.[8] To provide some coordination between these papers they will all be commented on here.

The zero-point spin deviation was measured by a double-resonance technique; the results are given in Ref. 5. In 3D systems the predicted deviations are small ($\sim 2\%$) for the best investigated system MnF_2. The uncertainties in the value of the super-transferred hyperfine coupling constant of the ^{55}Mn nuclei in the ordered system[9] are such as to make comparison of theory and experiment marginal. In the QL structure, on the other hand, simple spin-wave theory predicts a reduction in $\langle S^z\rangle$ of ≈ 0.2 and this is substantially greater than in 3D cases. The results found in Refs. 5 and 6 gave values of 0.20 ± 0.03 for K_2NiF_4 and 0.17 ± 0.03 for K_2MnF_4 and Rb_2MnF_4, in very good agreement with the values predicted by spin-wave theory including the anisotropy. Results from various perturbation calculations give substantially lower values.[10] It is not really clear why the agreement with spin-wave theory is so good. Stinchcombe[11] has calculated S^{-1} corrections to spin-wave theory which go to zero as the anisotropy vanishes and adduces this as a justification for the good result of the S^0 theory, since in the present case the anisotropy is small. However, it does not seem likely that when the anisotropy goes to zero, spin-wave theory will become exact and it would appear necessary to calculate the S^{-2} corrections before accepting the argument.

The variation of sublattice magnetization with temperature is measured in the conventional way by following the NMR of suitable F nuclei as the temperature is varied. The results show clearly that spin-wave theory in its simplest form gives an excellent account of the magnetization up to about $0.4T_N$. The two-dimensional nature of the magnetic ordering, which has a marked effect on the whole functional form of the $\langle S^z\rangle$-vs-T curve is thus confirmed. Attempts to extend the fit to higher temperatures by simple renormalization procedures, which are described in detail in the current paper, are not markedly successful. A meth-

od employed by Low for MnF_2[12] was used. This renormalizes the spin-wave energies by a term involving occupation numbers to essentially order S^{-1}. The occupation numbers themselves are then found self-consistently. (The spin-wave gap is inserted into the calculation as a known quantity.) The treatment has a slightly hybrid character, but the nature of the results seems to suggest that no simple renormalization scheme is likely to be effective. In K_2MnF_4 and Rb_2MnF_4 the region of good fit is extended by about 3 K, while in K_2NiF_4 no improvement whatsoever is found. At the same time the values of the exchange constants and spin-wave gap energies derived from the fits are in good agreement with other estimates. Also the renormalization dispersion relation which can be deduced agrees very well with that of recent neutron measurements on K_2MnF_4.[8]

The failure of the modified spin-wave theories to describe the behavior of the magnetization takes place in K_2NiF_4 when only a 4% extra thermal deviation has been added to the 18% of the zero-point motion. In K_2MnF_4 and Rb_2MnF_4 it occurs when 7% thermal has been added to 7% zero point. On the other hand, in 3D, Low found substantial agreement up to a deviation of 2% zero point plus 50% thermal. It is clear that in the 2D systems, when the spin-wave theory starts to underestimate the fall of the sublattice magnetization, there has not been a prolific excitation of magnons and occupation number renormalizations are not significant. Presumably one is then starting to see the contribution of excitation processes which are characteristic of the transition or critical region. All available evidence from neutron scattering,[2,8] the linewidth of antiferromagnetic resonance[7] and that of EPR,[13] indicates that in 2D systems such processes extend over a wider temperature range than in 3D ones, both above and below the critical temperature.

It was mentioned above that in attempting to fit the sublattice-magnetization data the temperature dependence of the energy gap was inserted into the calculations independently. This energy gap is just the antiferromagnetic resonance frequency and is, in principle, accessible to direct determination. In K_2NiF_4 it lies in an experimentally convenient region and has been measured by Birgeneau, DeRosa, and Guggenheim.[14] For K_2MnF_4 and Rb_2MnF_4 the direct measurement is not readily available in zero field. In a field of ≈ 40 kOe the gap may be reduced to microwave frequencies. The measurement of the field for resonance at 24 GHz is described in Ref. 7. It is possible to deduce from these observations the gap in zero field by extending the Oguchi-type renormalization method already cited to cases where a magnetic field is present. The system parameters used in this reduction are themselves found from the fitting of the sublattice magnetization.

II. SPIN-WAVE THEORY

In this section we review the results of spin-wave theory necessary to our discussion of sublattice magnetization in the QL structure, and note the assumptions upon which our interpretation is based. This material is found in many sources; an excellent general reference is the review by Keffer.[15]

Sublattice magnetization calculations for the QL compounds are based on a model of nearest-neighbor isotropic exchange coupling J, with anisotropy represented as a temperature-dependent staggered field H_A at the sites of the atomic spins. Thus, the Hamiltonian is given by

$$\mathcal{H} = |J| \sum_{\langle l,m \rangle} \vec{S}_l \cdot \vec{S}_m - g\mu_B H_A \left(\sum_l S_l^z - \sum_m S_m^z \right) , \quad (1)$$

where l and m are summed over the two magnetic sublattices, respectively. Our first goal is to summarize the calculations of $\langle S^z \rangle$ for a strictly 2D system using simple spin-wave theory and the lowest-order corrections to this picture developed by Oguchi.[16] Small corrections to this approximate picture—including interactions with more distant neighbors, effects of dipolar anisotropy, and interactions with neighboring layers—will be considered in due course.

In zero applied field the model situation described above has two degenerate spin-wave branches with energies given by[16]

$$E_{\vec{k}}/4|J|S = [(1+\alpha)^2 - \gamma_{\vec{k}}^2]^{1/2} - g_{\vec{k}}[R_0 + R_1(T)] , \quad (2)$$

with

$$g_{\vec{k}} = (1 + \alpha - \gamma_{\vec{k}}^2)\,[(1+\alpha)^2 - \gamma_{\vec{k}}^2]^{-1/2}, \quad (3)$$

$$R_0 = (1/2SN_0) \sum_{\vec{k}} (g_{\vec{k}} - 1) , \quad (4)$$

and

$$R_1 = (1/2SN_0) \sum_{\vec{k}} g_{\vec{k}} (n_{\vec{k}}^{(1)} + n_{\vec{k}}^{(2)}) , \quad (5)$$

where $\alpha = g\mu_B H_A/4|J|S$ is the anisotropy parameter, $n_{\vec{k}}^{(1)} = n_{\vec{k}}^{(2)} = [e^{\beta E_{\vec{k}}} - 1]^{-1}$ are the Bose occupation numbers for the two spin-wave branches, and $\gamma_{\vec{k}} = \cos(\frac{1}{2}k_x a)\cos(\frac{1}{2}k_y a)$ for the QL structure. N_0 is the number of magnetic unit cells in the system. The correction terms in Eq. (2) have also been derived by Keffer[15] using physical arguments. The quantities R_0 and R_1 are first-order corrections in the expansion parameter in $1/S$. One objective in this study is to see what improvement in fitting to the experimental sublattice magnetization data can be effected by including these terms in the analysis.

R_0 is a negative definite quantity so that $-g_{\vec{k}}R_0$ gives a temperature-independent fractional in-

crease in magnon energy varying from $\frac{1}{2}R_0$ at the bottom of the band to $R_0/(1+\alpha)$ at the zone boundary. As discussed previously,[17] however, this correction is very nearly "invisible" to experiments because its effect is simulated to within parts in 10^4 by adjusted values of J and α in the zero-order dispersion relation. Thus, at low temperatures $[R_1(T)\approx 0]$, Eq. (2) is closely approximated by the dispersion relation of simple spin-wave theory,

$$E_{\vec{k}}/4|J_s|S=[(1+\alpha_s)^2-\gamma_{\vec{k}}^2]^{1/2} \ , \tag{6}$$

where we identify the equivalent simple spin-wave-theory parameters throughout with the subscript s. J_s and α_s are obtained by equating Eqs. (2) and (6) at the zone boundary and at $k=0$, which yields

$$J_s(1+\alpha_s)=J(1+\alpha-R_0) \tag{7}$$

and

$$J_s(2\alpha_s+\alpha_s^2)^{1/2}=J(2\alpha+\alpha^2)^{1/2}[1-R_0/(2+\alpha)] \ . \tag{8}$$

For α_s, $|R_0|\ll 1$, these yield $J_s\approx J(1-R_0)$ and $\alpha_s \approx \alpha/(1-R_0)$.

The temperature-dependent renormalization term $-g_{\vec{k}}R_1(T)$ acts to lower the magnon energies as the temperature is raised. This term was found by Low[12] to bring about a markedly improved agreement with sublattice magnetization data for MnF_2. The gap temperature $T_G(T)=E_{k=0}/k_B$ is dependent on R_1, as expressed by the general equation [Eq. (2) with $k=0$]

$$T_G(T)=(4|J|S/k_B)\{(2\alpha+\alpha^2)^{1/2} -[R_0+R_1(T)]\,\alpha/(2\alpha+\alpha^2)^{1/2}\} \ . \tag{9}$$

It is usually found, however, that T_G decreases much more rapidly with T than Eq. (9) would permit by virtue of $R_1(T)$ alone. We have therefore adopted the *experimental* variation of $T_G(T)$ with temperature by allowing α to be temperature dependent. In fact, we have set Eq. (9) equal to the experimentally determined $T_G(T)$ in order to obtain $\alpha(T)$.[18]

The average magnetization residing at any atomic site is given by $g\mu_B\langle S^z\rangle$, where

$$\langle S^z\rangle = S-\Delta_0-\Delta S(T) \ . \tag{10}$$

Here S is the spin multiplicity,

$$\Delta_0=\frac{1}{2N_0}\sum_{\vec{k}}\left(\frac{(1+\alpha)}{[(1+\alpha)^2-\gamma_{\vec{k}}^2]^{1/2}}-1\right) \tag{11}$$

is the zero-point spin reduction (note that Δ_0 is weakly temperature dependent through the parameter α), and $\Delta S(T)$ is the temperature-dependent part of $\langle S^z\rangle$ given by

$$\Delta S(T)=\frac{1}{2N_0}\sum_{\vec{k}}\frac{1+\alpha}{[(1+\alpha)^2-\gamma_{\vec{k}}^2]^{1/2}}\,(n_{\vec{k}}^{(1)}+n_{\vec{k}}^{(2)}) \ . \tag{12}$$

Computer evaluations of $\langle S^z\rangle$ will be discussed in the following sections. Here we only briefly mention the way in which, given $\alpha(T)$ and $|J|$, the calculation of $\langle S^z\rangle$ at a certain temperature was implemented on the computer. First, R_0 and Δ_0 are calculated by the summations Eq. (4) and (11) over the Brillouin zone. Subsequently, R_1 is calculated by iterating the self-consistent set of Eqs. (2) and (5). Then, all constants in the dispersion relation Eq. (2) are known, and the calculation is completed by inserting the dispersion relation into the Bose factors and evaluating Eq. (12). Evaluation of the summations over the Brillouin zone is in general quite involved in three dimensions. In two dimensions, however, the summations can be greatly facilitated when, on converting sums to integrals, use is made of the relation

$$N_0^{-1}\sum_{\vec{k}}F(\gamma_{\vec{k}})=(4/\pi^2)\int_0^1 dz\ K((1-z^2)^{1/2})F(z) \ . \tag{13}$$

Here $K(m)=\int_0^{\pi/2}dx[1-m^2\sin^2x]^{-1/2}$ is a complete elliptic integral of the first kind, for which fast computer subroutines are available.

Convenient low-temperature approximations to $\Delta S(T)$ have been discussed in earlier publications,[6,17] especially in connection with the K_2NiF_4 data. We mention them here for completeness. For $kT\ll 4|J|S$, one may integrate Eq. (12) in closed form by taking $\gamma_{\vec{k}}^2\approx 1-\frac{1}{4}k^2a^2$, yielding

$$\Delta S(T)\approx\frac{-(1+\alpha_s)k_BT}{2\pi|J_s|S}\ln(1-e^{-T_G/T}) \ . \tag{14}$$

In the case that second- and third-neighbor intralayer exchange interactions are important, the exchange constant J_s in Eq. (14) is modified to become $J_{1s}-2J_{2s}-4J_{3s}$ in an obvious notation. This is a long-wavelength approximation and should be valid for the low-temperature studies conducted here.

In the above development we have assumed a $\vec{k}$-independent anisotropy field such as would be effectively provided, for example, by a uniaxial crystalline field. For the Mn^{2+} QL compounds, however, the major source of anisotropy is dipolar interactions. We have examined in detail the question of how this might affect the above treatment. Exact diagonalizations of the dipolar anisotropy case are available,[19] and are discussed in Appendix A. With these one can assess the accuracy of the dispersion relation Eq. (2) as an approximation to the dipolar case. The details are relegated to Appendix A, with the general conclusions as follows. The dipolar interactions lift the degeneracy of the two spin-wave branches and, in addition, cause their average value to deviate from the form of Eq. (2). For $\alpha\approx 0.004$ (i.e., for K_2MnF_4 and Rb_2MnF_4) the latter effect is of the order of 0.1% or less throughout the first Brillouin zone, and is

therefore negligible for our purposes. Moreover, the mode splitting, which ranges up to 1% of E_g, is expected to cancel to a high degree in the thermodynamic averages. This splitting vanishes as $k \to 0$; thus, there is no degenerate manifold of excitations near $k = 0$ as is found in three dimensions.[20]

Lastly, we consider the effect on our analysis of possible three-dimensional effects, i.e., interactions with spins in nearby layers of the QL structure. It was noted by Legrande and Plumier[21] that the coupling between adjacent layers, about 7 Å apart in these compounds, cancels at long wavelengths because of the staggered registry of the sublattices. In addition, neutron studies of the c-axis magnon dispersion in K_2NiF_4[22] and EPR results[23] for dilute $K_2MgF_4:Mn^{2+}$ show that these nearest-neighbor interlayer couplings are below 1% of the primary exchange interactions. They will not be considered further.

Of potentially greater importance, in spite of their minuscule size, the couplings between second-neighbor layers. Their effect on the magnon dispersion relation is investigated in detail in Appendix B for both ferromagnetic and antiferromagnetic ordering in the third dimension. Both such phases have been observed experimentally.[2] In a model calculation assuming exchange coupling J_c between corresponding moments in second-neighbor layers, a fractionally small modulation of the magnon energy is found under the assumption $|J_c/J| \ll 1$. The effect on the sublattice magnetization is found to be well approximated by a modified gap temperature.

$$T_G \to T_G[1 + |J_c| S/(g\mu_B H_A)] \ , \qquad (15)$$

where it is noted that J_c need only become comparable to H_A in order for its effects to be felt. The present experiments are sensitive to J_c values as small as $|J_c/J| \approx 10^{-4}$ because of the weak anisotropy. In contrast, J_c alters the $k = 0$ AFMR mode only by amounts of order $|J_c/J|$; thus, it is the discrepancy between T_G as measured by AFMR and by fitting the sublattice magnetization which reveals any appreciable c-axis coupling. In Secs. IV and V the experimental results are used to establish upper limits for $|J_c|$ in the QL compounds under investigation. The dipolar contribution to J_c is of order $10^{-4}\, g\mu_B H_A$ in the Mn^{2+} compounds and smaller in K_2NiF_4.

III. EXPERIMENTAL

The temperature dependence of the sublattice magnetization has been measured by monitoring the nuclear-magnetic-resonance (NMR) frequency $f(T)$ of the out-of-layer ^{19}F nuclei located adjacent to the Mn ions. The external field being zero, these nuclei resonate exclusively in a transferred hyperfine field. That is, $f(T) = A_{19}\langle S^z \rangle$, where A_{19} is the hyperfine coupling constant which results from transferred hyperfine interaction with an additional contribution ($\approx 2\%$) from dipolar origin. The results are presented in Fig. 1 for K_2NiF_4,[6] and in Fig. 2 for K_2MnF_4 and Rb_2MnF_4.[24] Data for K_2MnF_4, presented by us previously,[6] have been carefully reexamined and partially repeated in the temperature region below 20 K. The samples were prepared in the way described earlier.[25]

The NMR technique used consists of exciting the nuclear precession by a rf pulse with duration of about 2 μsec, and observing the free-induction decay following the pulse. The coil around the sample was used for both excitation and detection. The detection system consisted of a broadband rf amplifier, with fast recovery after overload, followed by a video detector. The frequencies were measured by beating the free-induction-decay signals with a standard oscillator, which was phase locked to the transmitter and weakly coupled to the detection system.

Generally speaking, the free-induction-decay times were of the order of 10 μsec for all samples at the lower temperatures, and decreased only

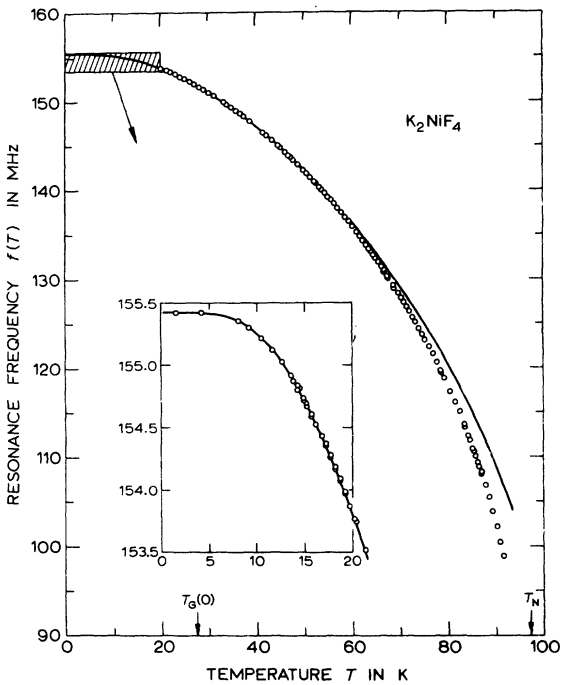

FIG. 1. NMR frequency $f(T)$ of the ^{19}F out-of-layer nuclei in K_2NiF_4 vs temperature T at zero external field. The spin-wave energy gap and the Néel temperature are indicated by $T_G(0)$ and T_N, respectively. The solid curve is calculated from renormalized spin-wave theory as described in the text with the output values of the least-squares fit up to 40 K.

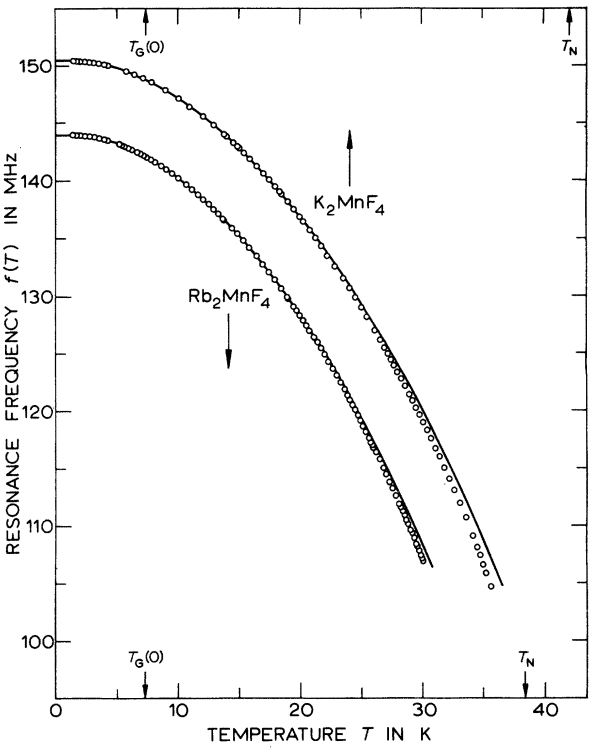

FIG. 2. Same as Fig. 1, but for K_2MnF_4 and Rb_2MnF_4. For the solid curves parameter values for the fits up to 18 K and 17 K, respectively, have been used.

slowly upon increasing the temperature. For K_2MnF_4 at 1.5 K, the limit of pure Suhl–Nakamura[26] linewidth (~ 30 kHz) was very nearly reached.[27] The Rb_2MnF_4 samples were unfortunately contaminated to the extent that about 1% of the Rb ions were replaced by K. This resulted in inhomogeneous broadening of the NMR line of the order of 50–100 kHz at the higher temperatures,[28] which is the reason for the fact that the results for Rb_2MnF_4 are less accurate than for K_2MnF_4. For K_2NiF_4 at 1.5 K we observed the spin-lattice relaxation time T_1 to be of the order of 10 min; it dropped rapidly to milliseconds as soon as temperatures corresponding to the spin-wave gap were reached. In K_2MnF_4 and Rb_2MnF_4 we observed $T_1 \approx 10$ sec at 1.5 K, apparently impurity dominated.

Temperature control was done below 4.2 K by immersion in liquid helium, between 13.8 and 20.3 K by immersion in liquid equilibrium hydrogen, and in the remaining regions by cooling in a continuous stream of boiled-off helium gas, the temperature of which was servostabilized. For K_2NiF_4 a series of data was also taken in the liquid-nitrogen region. Temperatures were measured by monitoring the pressure above the liquid coolants, with hydrostatic correction when necessary, or with reference to a standard platinum resistor and calibrated germanium resistor thermometers. All thermometers were carefully checked at the boiling point of helium, the triple and boiling points of equilibrium hydrogen, and the triple and boiling points of nitrogen.[29] The accuracy of the temperature measurements is estimated to be 0.5% or 0.2 K, whichever is less. In the region between 4.2 and 13.8 K, however, the error limit is increased to 0.1 K. In the over-all errors of the data points, which have been entered into the least-squares fits (cf. Figs. 5, 6, and 7, to be discussed below), we have combined the estimated errors in frequency and temperature measurements.

A remark should be made on the temperature dependence of the hyperfine constant A_{19}, which we have taken to be constant throughout the temperature region of interest. In general, the transferred hyperfine interaction will be sensitive to the Mn^{2+}-F^- separation, and is therefore dependent on lattice vibrations and thermal expansion. No study of these effects has been made in the QL compounds as yet, but a similar situation is encountered in the system of Mn^{2+}-doped LiF, which has been discussed by Shrivastava.[30] The effect of harmonic lattice vibrations is shown to be proportional to T^4 at low temperatures. The thermal dilatation will follow the trend of the internal thermal energy,[31] which also will fall off with a T^4-type dependence. From high-pressure data[32] Shrivastava estimated thermal expansion to account for about one-half of the decrement of A_{19} with temperature, while the other half was shown to have a fractional change given by $K_s T^4$ with $K_s = 1.5 \times 10^{-12}$ K^{-4}. At say 25 K, the highest temperatures used in the discussion of spin-wave theory for K_2MnF_4 and Rb_2MnF_4, this implies a fractional decrease of A_{19} by 5×10^{-7}, or 0.08 kHz in the NMR frequencies. This estimate may be too small by an order of magnitude or so because the Debye temperature may be lower in the layer compounds, but anyway the effect will be far below the the experimental errors from other sources. For K_2NiF_4, spin-wave fits (Sec. IV) appear to be possible up to 40 K, where with allowance for difference in Debye temperature there results a correction of the order of 20 kHz. Another way of estimating the effect in the case of K_2NiF_4 is from comparison of A_{19} as measured here at $T \approx 0$ K ($A_{19}/h = 187$ MHz with $\Delta_0 = 0.18$) with the value at 297 K ($A_{19}/h = 133$ MHz).[33] Accounting for the change in the dipolar part in going from the antiferromagnetic to the paramagnetic state, a decrease of A_{19} by 28% is found over 300 K,[34] which implies a change of 2×10^{-4} over the first 50 K, corresponding to a correction of 30 kHz in the NMR frequency. For further discussion, see Sec. IV.

IV. K_2NiF_4 IN SPIN-WAVE REGION

A good deal of detailed information is available regarding the magnetic interaction parameters in

K_2NiF_4. We review these data briefly before discussing the sublattice magnetization analysis.

Neutron scattering measurements of the spin-wave dispersion at low temperatures[22] give

$$[J_{1s} - (J_{2s} + 2J_{3s})]/k_B = -112.3 \pm 0.3 \text{ K}.$$

Data on the second- and third-neighbor couplings have been reported[23,35] for dilute $K_2MgF_4 : Ni^{2+}$, giving $J_2/k_B = -0.6$ K, with J_3 an order of magnitude smaller. In view of the good agreement between J_1 measured earlier for these systems[35] and the neutron value for K_2NiF_4, we assume J_2 and J_3 are also correct for the concentrated antiferromagnet. The value of the single exchange parameter $J_s = J_{1s} - 2(J_{2s} + 2J_{3s})$ relevant to the sublattice magnetization at low temperatures [see Eq. (14)] is then taken to be -111.7 ± 0.6 K. The corresponding uncorrected exchange constant is $J/k_B = -103.5 \pm 0.6$ K. Combining J and the measured 4.2 K AFMR splitting of 27.48 K ± 1% with $g = 2.22$,[14] we find, using Eq. (9), $\alpha = 0.0021$ and $H_A \approx 5.73$ kG. This is nearly an order of magnitude larger than the dipolar field (≈ 1.3 kG) and is therefore due primarily to single-ion and anisotropic exchange effects. Yamaguchi[33] finds (again, for $K_2MgF_4 : Ni^{2+}$) a single-ion anisotropy term $\mathcal{H}_{\text{anis}} = DS_z^2$ with $D/hc = -0.425$ cm^{-1}, giving $H_{A\,\text{single ion}} = 4.1$ kG. No data are available on the nearest-neighbor anisotropic exchange coefficient to compare with D. It appears, however, that D is the primary source of anisotropy in K_2NiF_4.

In analyzing the data we shall assume the spin-wave energy gap to scale with the sublattice magnetization, $T_G(T) \propto \langle S^z \rangle$, as has been observed experimentally.[14] There are then essentially three parameters required to fit the ^{19}F NMR frequency data to

$$f(T) = f(0)\langle S^z \rangle / \langle S^z \rangle_{T=0} \ , \qquad (16)$$

with $\langle S^z \rangle$ calculated from the renormalized spin-wave theory in the way outlined in Sec. II. These parameters are J, the zero-temperature spin-wave energy gap $T_G(0)$, and the zero-temperature frequency $f(0)$. A series of least-squares fits was carried out over temperature ranges extending from 1.5 K up to a selection of upper limits. In doing this the integrals over the Brillouin zone were evaluated at the experimental temperatures of the data points with estimates of J and $T_G(0)$, while further adjustment of $\Delta S(T)$ was carried out by means of the approximate formula Eq. (14). This process was repeated to ensure self-consistency.

Results of the fitting for J and $T_G(0)$ are plotted in Fig. 3, where the closed circles represent parameter values obtained in data fits including all data up to the temperatures indicated. The variation of corresponding output values of $f(0)$ is well within the errors, and further, the output values for $f(0)$ appear to be only weakly correlated with those for J and $T_G(0)$. We therefore give $f(0)$ as a figure entry in Fig. 3, and do not consider it further. The points in Fig. 3 are somewhat scattered but tend to lie along a straight line because the errors in J and $T_G(0)$ are highly correlated. That this is so may be seen by plotting constant error contours in the $[J, T_G(0)]$ plane, with standard deviations and correlation parameters propagated from the errors in the measured NMR frequencies by the least-squares-fitting program. The contour for an excursion of two standard deviations from the least-squares-fitted values of J and $T_G(0)$, which has a 98% probability of enclosing the true values, is an ellipse given by

$$\{\delta J^2 + [\delta T_G(0)]^2 - 2\sigma\, \delta J\, \delta T_G(0)\}/(1-\sigma^2) = 4 \ , \qquad (17)$$

where δJ and $\delta T_G(0)$ are the variations of J and $T_G(0)$ in units of their standard deviations, and σ is the correlation parameter. The ellipse for all data up to 25 K (Fig. 3), i.e., up to a temperature comparable to the spin-wave gap, illustrates the expansion of error limits in a case of extreme

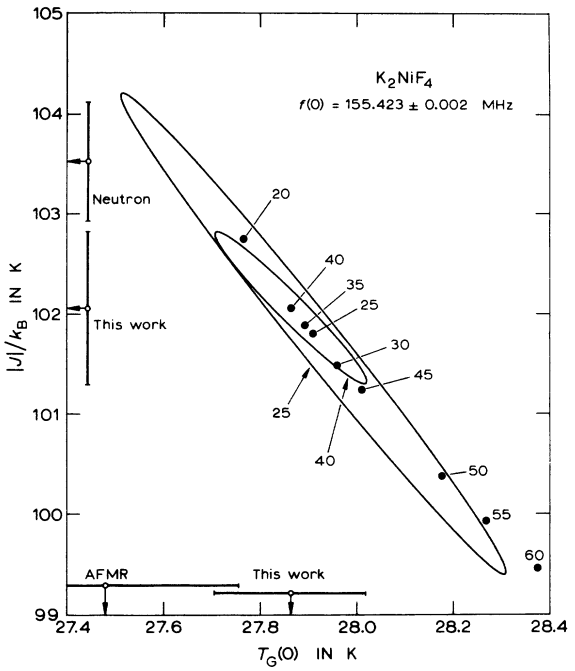

FIG. 3. Output values of the least-squares fits of the NMR frequency $f(T)$ in K_2NiF_4 to renormalized spin-wave theory. The closed circles represent the output values for the exchange integral J and the spin-wave gap temperature $T_G(0)$ obtained from fits to all data up to the temperature indicated. Error ellipses of two standard deviations are given for the fits up to 25 K and 40 K only. The output values of the 40-K fit, projections of the 40-K ellipse on the axes, are marked "this work." The value for J from neutron diffraction has been derived from J_s in Ref. 22, the AFMR value of $T_G(0)$ from Ref. 14.

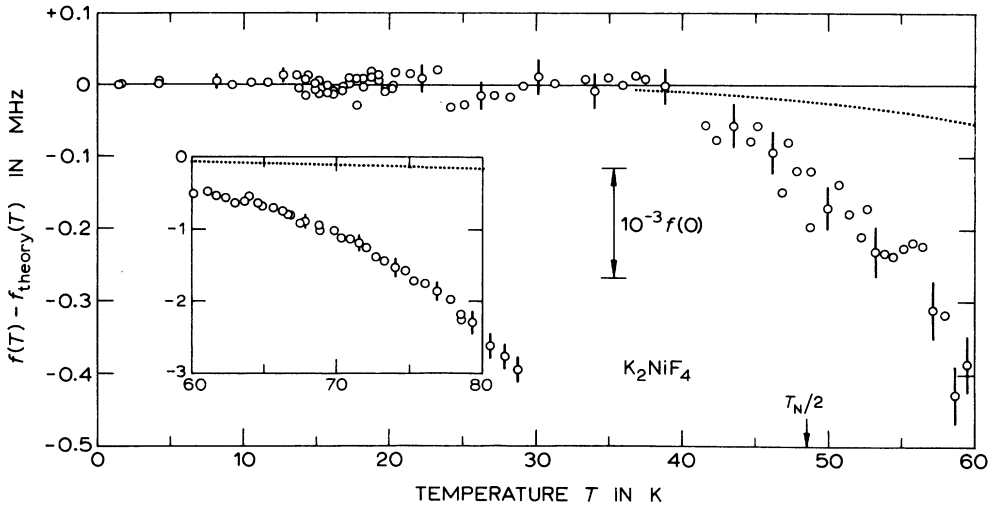

FIG. 4. Deviation of the experimental resonance frequency $f(T)$ in K_2NiF_4 from the frequency calculated from renormalized spin-wave theory with the parameters obtained in the 40-K least-squares fit, showing a distinct breakdown of the theory above this temperature. The estimated effects of a reduction of A_{19} with temperature are indicated by the dotted line.

correlation ($\sigma = -0.99$). On the other hand, the minor axis coordinates of these ellipses are determined with high precision. As one might expect, the parameter points are scattered along the major axis of this ellipse at temperatures up to the breakdown of the spin-wave fit, after which there is a distinct trend to move downward to the right. A second ellipse is plotted for all data up to 40 K, and is thought to give the most precise determination of J and $T_G(0)$ of the series. Here the correlation is somewhat smaller ($\sigma = -0.97$) and the errors considerably reduced. A plot of the deviations of the experimental data from theory is given for this fit in Fig. 4. The agreement with theory is excellent up to 40 K, with a distinct breakaway above that point. The least-squares program was also able to fit the data to 45 K, but not beyond, within the experimental errors. However, the downward trend above 40 K is also clearly visible in that case, giving a definite indication of the breakdown of the spin-wave fit.

The spin-wave theory fitted to the K_2NiF_4 data included the Oguchi renormalization terms in line with the discussion of K_2MnF_4 and Rb_2MnF_4 in Sec. V following. It should however be noted that in the case of K_2NiF_4, spin-wave theory without these renormalization terms will do equally well. The temperature-dependent renormalization is negligibly small throughout the range of the spin-wave fit ($R_1 \lesssim 0.001$). Although $R_0 = -0.079$ is quite large, the resulting modification of the form of $E_{\vec{k}}$ cannot be resolved within the accuracy of the data.

In Sec. III it was pointed out that at temperatures of 50 K and higher, A_{19} may be reduced. In Fig. 4 the effects of such a reduction are indicated by the dotted line, which represents the spin-wave fit, but with the inclusion of an estimated fractional decrement of A_{19} by $K_s T^4$ with $K_s = 3.0 \times 10^{-11}$ K^{-4}. The reduction of A_{19} possibly could increase the fit to spin-wave theory to slightly higher temperatures, but, as in seen in Fig. 4, is unlikely to extend the upper limit of the fit to beyond, say, 50 K.

It is interesting to compare the parameter determination of the 40-K fit with values determined by other techniques. The latter values with error limits are shown in the margins of Fig. 3. The neutron value[21] for J_s, corrected using Eqs. (7) and (8) to obtain J, gives $|J|/k_B = 103.5 \pm 0.6$ K. This is somewhat larger than the 40 K fitted value 102.1 ± 0.8 K. Although this discrepancy is only marginally resolved, it may reflect a slightly larger value of Δ_0 than the spin-wave value ($\Delta_0 = 0.177$ for $\alpha = 0.0021$) taken in the calculation. We recall that in the case of K_2NiF_4, with a zone-boundary spin-wave energy of ≈ 450 K, the k^2 approximation leading to Eq. (14) is quite a good approximation. From inspection of Eqs. (14) and (16) it follows that, if Δ_0 is allowed to vary in the fit, one can only determine the value of the product $J(S - \Delta_0)$. As in a previous publication,[6] $J(S - \Delta_0)$ is combined with the neutron determination of J to arrive at a value of Δ_0 consistent with both experiments, with the result $\Delta_0 = 0.193 - 0.81(J_{2s} + 2J_{3s})/J_{1s} \pm 0.02 = 0.19 \pm 0.02$. The zero-temperature gap energy $T_G(0)$ is found to be in marginal agreement with the AFMR value 27.48 K $\pm$ 1%.[14] Thus there may still be a c-axis exchange effect (see Appendix B), but it is not well resolved. The observed dis-

crepancy in gap energies leads to $|J_c/J| \sim 1.2\times10^{-4}$.

V. Mn^{2+} COMPOUNDS IN SPIN-WAVE REGION

The application of spin-wave theory to the sublattice magnetization data for these compounds was carried out in a fashion similar to the K_2NiF_4 case. There are, however, a number of differences in detail. Use has been made of the AFMR measurements for these compounds reported in the following paper.[7] These measurements have been interpreted with a renormalization formalism which reduces to that of Sec. II for zero applied field, and, given J, yields the spin-wave energy gap as a function of temperature. In order to allow for possible c-axis exchange modification of T_G (see Appendix B), however, in the least-squares fitting only the *relative* variation of the experimental T_G with the temperature has been retained, while the gap at zero temperature $T_G(0)$ has been taken as a parameter. As in the case of K_2NiF_4, $f(0)$ and J are also allowed to vary in the least-squares fitting. Although the energy gaps derived from the AFMR measurements are only weakly dependent on the value of J,[7] the output values of the fitting to the NMR data have been fed back to the analysis of the AFMR data for the sake of consistency.

As for K_2NiF_4 a neutron scattering study of the low-temperature magnon dispersion has been made for K_2MnF_4. These measurements, reported in the second following paper,[8] yield $J = 8.45 \pm 0.1$ K. Unfortunately, no such measurement has been reported for Rb_2MnF_4, nor are there any data on exchange couplings with more distant neighbors. Data for the closely related perovskites[36] suggest that J_2 is of order of 1% or so. There is no evidence for second-neighbor exchange in the presently reported dispersion measurements.[8] The zero-point spin reduction Δ_0 has been measured by use of a double-resonance technique[5] to be $\Delta_0 = 0.17 \pm 0.03$ for both K_2MnF_4 and Rb_2MnF_4. This in good accord with the spin-wave theory, Eq. (11), which yields $\Delta_0 = 0.170$ and 0.167, respectively, for appropriate values of α. We therefore henceforth adopt Eq. (11), noting that uncertainty in $S - \Delta_0$ makes some contribution to the absolute error in our determination of J.

A. K_2MnF_4

Values of J and $T_G(0)$ obtained in a series of least-squares fits to the NMR data up to the temperatures shown are plotted in Fig. 5. As before $f(0)$ is given as a figure entry. The fully renormalized theory, Eqs. (2)–(5), is employed here. Two error ellipses are plotted as described in Sec. IV for an excursion of two standard deviations. Again, we find using data up to the gap temperature region [$T_G(0) = 7.40 \pm 0.05$ K] large and highly correlated errors ($\sigma = -0.97$) in the output values of J and $T_G(0)$. Considerable reduction of the errors is obtained with the fit up to 18 K.

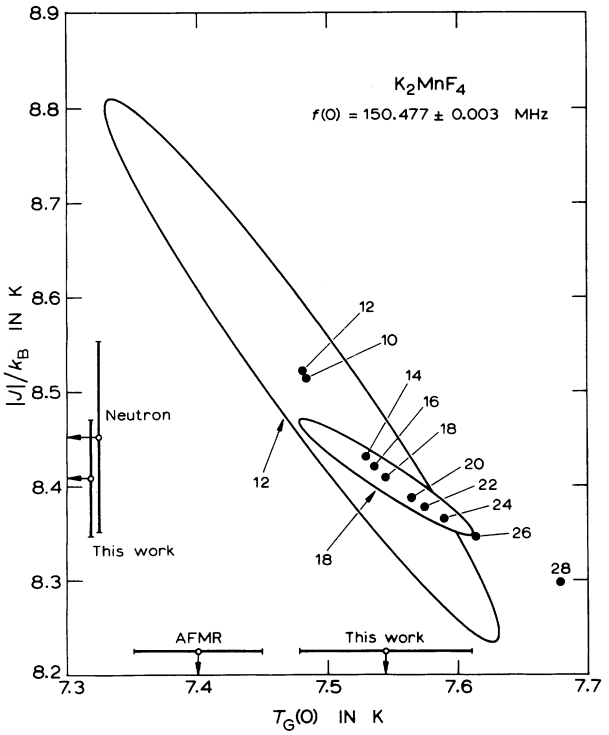

FIG. 5. Same as Fig. 3, but for K_2MnF_4. The most precise fit is the one for all data up to 18 K. The neutron value of J is derived from Ref. 8, and the AFMR value of $T_G(0)$ is taken from Ref. 7.

The least-squares value of J we find is seen to be in excellent agreement with the neutron value plotted,[8] as well as with the value $J = 8.4 \pm 0.1$ K derived from susceptibilities by Breed.[1] This agreement may be fortuitous because of the combined uncertainty in the zero-point reduction Δ_0 and second-neighbor exchange J_2. These results are consistent with the spin-wave value $\Delta_0 = 0.170$ and negligible J_2. The fitted value of $T_G(0)$ is ~2% larger than the AFMR value, a discrepancy which is only barely resolved. This difference is indicative of a c-axis exchange coupling, corresponding to $|J_c/J| \sim 3\times10^{-4}$ [Eq. (15)].

The deviations of the experimental data from the fitted theoretical curve are given in Fig. 6 for the 18-K fit to the K_2MnF_4 NMR data. For the fully renormalized theory (open circles) the fit is within the errors of the data up to 18 K. With a distinct downward trend thereafter. The "bump" between 5 and 13 K, within the experimental errors, we attribute to thermometry errors. The most accurate data are in the liquid-helium and hydrogen temperature intervals, the parameters of the fit being most stringently determined by the latter re-

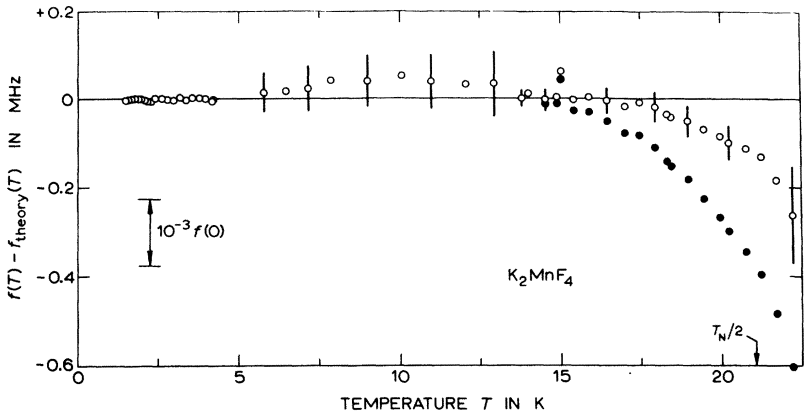

FIG. 6. The deviation of the experimental resonance frequency $f(T)$ in K_2MnF_4 from the frequency calculated from renormalized spin-wave theory (open circles) with the parameters obtained in the 18-K least-squares fit. A similar calculation for the unrenormalized theory (closed circles) with J and α from the 18-K renormalized fit [adjusted according to Eqs. (7) and (8)] shows that renormalization extends the range of the fit by only 3 K.

gion.

One objective of the present work was to test the role of the temperature-dependent renormalization factor R_1 in improving the agreement of spin-wave theory with the sublattice magnetization data. To this end an unrenormalized calculation of $\Delta S(T)$ was carried out using the parameters from the 18-K fit above by omitting the effect of R_1. The results are presented as the lower points (closed circles) in Fig. 6. The difference between the two calculations is negligible below 13 K. Thus, Fig. 6 illustrates the effects of temperature-dependent renormalization in fitting the sublattice magnetization data. The improvement is not substantial, extending the range of fit by only ~3 K, to a temperature not yet one-half the Néel temperature (T_N = 42.14 K).[37]

B. Rb_2MnF_4

The results for this compound are quite similar to those for the potassium isomorph. Parameter values obtained in a series of fits to the renormalized theory, Eqs. (2)–(5), are given in Fig. 7. Again, we plot two error ellipses, one for all data up to a temperature near the magnon gap value [$T_G(0) = 7.28 \pm 0.05$ K] and one for 17 K, which we consider to be the upper limit of the spin-wave fit. These exhibit the same decreases of correlation and output errors with increasing temperature as found for the other compounds. The errors at 17 K are slightly larger than for K_2MnF_4, reflecting somewhat more scatter in the measured frequencies. This, in turn, is attributed to the potassium impurity problem mentioned in Sec. III.[28] $T_G(0)$ is somewhat larger than the AFMR value, as in the case of K_2MnF_4, with a barely resolved discrepancy which gives $|J_c/J| \sim 4\times10^{-4}$. The fitted value of J is slightly larger than, but essentially consistent with, those deduced from susceptibilities by Breed.[1]

A graph, analogous to Fig. 6, is given in Fig. 8 for the 17-K fit to Rb_2MnF_4. The general character of these plots is rather similar with somewhat increased data scatter clearly evident for Rb_2MnF_4. Again, the fit extends up to about $\frac{1}{2}T_N$ (T_N = 38.4 K) and the withdrawal of renormalization is found to diminish the range of the fit by a few degrees.

As expected, the two Mn^{2+} isomorphs are seen to behave in closely similar fashion, the main difference being a ≈15% smaller exchange coupling for the Rb compound. There are two other results which seem inconsistent with the ratio of exchange constants found here. First, we may deduce values for the anisotropy fields in these materials from the formula $H_A \approx 4|J|S\alpha/g\mu_B$, where α is obtained from the AFMR studies[7] and J is taken from

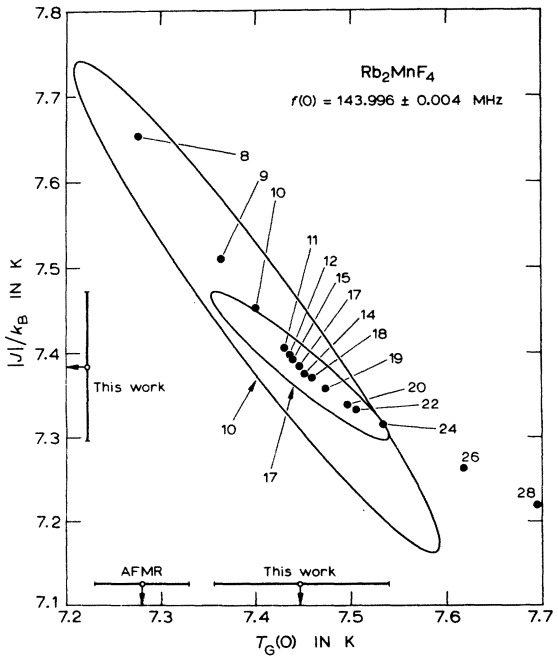

FIG. 7. Same as Fig. 3, but for Rb_2MnF_4. The most precise fit is the one for all data up to 17 K. The AFMR value of $T_G(0)$ is taken from Ref. 7.

the results of this section. These may then be compared with calculated dipolar values as set forth in Appendix A. The results are listed in Table I. The calculated dipolar values are based on unit-cell dimensions reported by Loopstra *et al.*[1] The deduced anisotropy field is seen to be about 200 G smaller than the dipolar value for K_2MnF_4, whereas it is found to be somewhat larger than the dipolar value for Rb_2MnF_4. These small discrepancies are attributed to uniaxial crystalline fields. Although the latter are of reasonable magnitude as indicated by electron-nuclear-double-resonance (ENDOR) measurements[38] on dilute $K_2MgF_4:Mn^{2+}$, the reversal of sign is perhaps somewhat unexpected for two compounds so similar in other respects.

A second, perhaps more curious discrepancy is found when we compare the ratio of exchange constants with the corresponding ratio of Néel temperatures. The Stanley-Kaplan[4] estimate of transition temperatures gives a reasonably good account of the T_N values for these materials, as is shown in Table I. However, one might expect that T_N scales with J for the two Mn compounds, which have only slightly different anisotropy. Instead, the Stanley–Kaplan estimate is 0.5 K below the observed value for K_2MnF_4 and 1.9 K below it for Rb_2MnF_4, a discrepancy which is outside the suggested errors.

VI. CONCLUSIONS AND DISCUSSIONS

The spin-wave and NMR parameters obtained in this and related studies of the QL compounds investigated here are summarized in Table I. For a detailed discussion of these results we of course refer the reader to Secs. IV and V. In general, there is satisfactory agreement between the exchange parameters determined here by adjusting spin-wave theory to fit the sublattice magnetization (NMR) data and those obtained from neutron scattering measurements. It is important to recall that the "fitted" J values may be "adjusted" over a range of one or two percent according to the prescription $J(S-\Delta_0)=\text{const}$ without appreciably altering the quality of the fit. Thus, one may bring our values of J into coincidence with the neutron ones in exchange for Δ_0's which are slightly larger than, but within experimental error of, the assumed spin-wave values.

The spin-wave gap energies reflected by these data lie above the measured AFMR values, with marginally resolved discrepancies in the direction which suggests minute exchange couplings with second-neighbor quadratic layers. Corresponding values are given for the effective couplings (J_c) between corresponding ions. A second magnetic phase has recently been observed[39] in K_2MnF_4 with a considerably higher transition temperature. This phase has been attributed[39] to Ca_2MnO_4-like ordering (antiferromagnetic) along the c axis. In view of the smallness of J_c, however, this would not appear to be an adequate explanation of the difference in T_N's.

For the QL compounds spin-wave theory has been found to give a good account of the magnon dispersion, zero-point reduction, low-temperature variation of sublattice magnetization, and spin-wave renormalization with temperature at all points in the zone except near $k=0$. The most surprising and interesting result is that the spin-wave description of the sublattice magnetization breaks down in all three compounds at temperatures less than $\frac{1}{2}T_N$ even with magnon energies *throughout the entire zone* correctly renormalized with temperature. The $k=0$ mode is renormalized according to experiment; nonzero $\vec{k}$ values are renormalized with the corrections developed by Oguchi[16] and confirmed by experiment for K_2NiF_4 (Ref. 2) and K_2MnF_4.[8]

It is also noteworthy that deviations in $\langle S^z\rangle$ from the zero-temperature value are unusually small at the point of spin-wave breakdown. For K_2NiF_4 the deviation is already nearly 20% from the Néel-state value $\langle S^z\rangle = S$ at $T=0$; at the point at which theory fails (≈ 40 K, see Fig. 4), $\langle S^z\rangle$ has only dropped

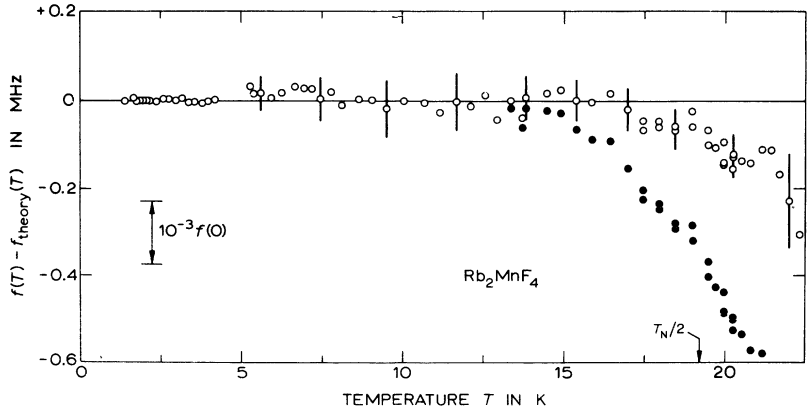

FIG. 8. Same as Fig. 6, but for the 17-K fit for Rb_2MnF_4. As in K_2MnF_4, renormalization does not extend the range of the fit by more than 3 K.

TABLE I. Summary of various quantities for the QL antiferromagnets K_2NiF_4, K_2MnF_4, and Rb_2MnF_4. Unless indicated otherwise, the values refer to the present work.

Parameter	K_2NiF_4	K_2MnF_4	Rb_2MnF_4
T_N (K)	97.1[a]	42.1[b]	38.4[a]
$T_{N,S-K}$ (K)	92[c]	41.6[c]	36.5[c]
Δ_0	0.177[d]	0.170[d]	0.167[d]
	0.19 ± 0.02[e]	0.17 ± 0.03[f]	0.17 ± 0.03[f]
R_0	−0.0790[g]	−0.0316[g]	−0.0316[g]
$f(0)$ (MHz)	155.423 ± 0.002	150.477 ± 0.003	143.996 ± 0.004
J/k_B (K)	−102.1 ± 0.8	−8.41 ± 0.06	−7.38 ± 0.09
	−103.5 ± 0.6[h]	−8.45 ± 0.1[i]	
		−8.4 ± 0.1[j]	−7.3 ± 0.1[j]
J_s/k_B (K)	−110.1 ± 0.8	−8.67 ± 0.06	−7.62 ± 0.09
	−111.7 ± 0.6[k]	−8.72 ± 0.1[l]	
$T_G(0)$ (K)	27.86 ± 0.16[m]	7.54 ± 0.07[m]	7.45 ± 0.09[m]
	27.48 ± 1%[n]	7.40 ± 0.05[o]	7.28 ± 0.05[o]
$\alpha(0)$	0.0021[p]	0.0038[p]	0.0047[p]
H_A (kG)	5.73[p]	2.35[p]	2.59[p]
$H_{A(D)}$ (kG)	1.28	2.57	2.48
$\lvert J_c/J\rvert$	$\leq 3\times10^{-4}$ [q]	$\leq 5\times10^{-4}$ [q]	$\leq 8\times10^{-4}$ [q]

[a]Reference 2.

[b]Reference 8.

[c]According to Stanley and Kaplan (Ref. 4) with this paper's J values.

[d]Spin-wave value, Eq. (11), assumed in the fitting procedure.

[e]Obtained by combining $J(S-\Delta_0)$ from this paper with J from neutron dispersion.

[f]Experimental value from a NMR-AFMR double resonance experiment (Ref. 5).

[g]Equation (4).

[h]From neutron dispersion measurements (Ref. 21), after correction for next-nearest-neighbor exchange and conversion from unrenormalized to renormalized spin-wave theory.

[i]From neutron dispersion (Ref. 8), after conversion to renormalized spin-wave theory.

[j]From perpendicular susceptibility at $T=0$ K (Ref. 1).

[k]From neutron dispersion (Ref. 21), after correction for next-nearest-neighbor exchange.

[l]From neutron dispersion (Ref. 8).

[m]This work; includes effects of residual c-axis exchange coupling.

[n]From AFMR (Ref. 14).

[o]From AFMR (Ref. 7).

[p]These values are derived from the AFMR results for the gap energy combined with J as obtained from the present work.

[q]The numbers quoted here are *upper limits* derived from the error estimates shown in Figs. 3, 5, and 7.

an additional 4% of S. For the Mn^{2+} cases the zero-point deviation is ≈ 7% of S with an additional 7% at the point of breakdown. These are in sharp contrast with MnF_2, where renormalized spin-wave theory[12] accounts for a deviation of ≈ 50% of S.

The above results suggest that at $T \approx \frac{1}{2}T_N$ and above, $\langle S^z \rangle$ for the QL compounds is partly determined by fluctuations in the z component of magnetization, i.e., "critical fluctuations," which are not treated by spin-wave theory. It is the presence of these other excitations, not the effects of spin-wave interactions, which causes the spin-wave description of $\langle S^z \rangle$ to fail at relatively low temperatures. Thus, no alternate formulation of spin-wave renormalization to the one employed in this paper[16] could be expected to improve matters. There is ample evidence for strong z-axis fluctuations in the QL compounds. Neutron scattering experiments on K_2NiF_4[2] reveal a large and unusual diffusive peak attributable to χ_{zz} at temperatures ranging well above and below T_N. In the Mn^{2+} isomorphs, the AFMR linewidth becomes extraordinarily large at and above the temperature where $\langle S^z \rangle$ deviates from the spin-wave description.[7] Such line broadening is very likely due to z-axis fluctuations. At the present time there is no theoretical formulation of these fluctuation modes with which to calculate their contribution to $\langle S^z \rangle$.

APPENDIX A: DIPOLAR ANISOTROPY IN TWO-DIMENSIONAL SPIN-WAVE THEORY

The effects of dipolar interactions on the antiferromagnetic spin-wave spectrum have been discussed by several authors, initially by Ziman,[19] who diagonalized that portion of the Hamiltonian which is quadratic in the spin-wave operators in the general case of an applied field with anisotropic g factor. We have reobtained this result for the case of nearest-neighbor exchange with zero applied field, giving for the eigenenergies

$$E_{\vec{k}}^{\pm}/4|J_s|S = \{1-\gamma_k^2 + (A+C-\gamma_k D+H_A)/2|J_s|S \pm [EE^* + 4\gamma_k^2 BB^* - 2\gamma_k(BE^*+B^*E)]^{1/2}/2|J_s|S\}^{1/2}, \tag{A1}$$

where (in the notation of Harris)[20]

$$\begin{aligned} A(\vec{k}) &= d\sum_n (3\cos^2\theta_n - 1)(1+\tfrac{1}{2}e^{i\vec{k}\cdot\vec{R}_n})/R_n^3 , \\ B(\vec{k}) &= -\tfrac{3}{4}d\sum_n \sin^2\theta_n e^{-2i\varphi_n} e^{i\vec{k}\cdot\vec{R}_n}/R_n^3 , \\ C(\vec{k}) &= -d\sum_n{}' (3\cos^2\theta_n - 1)/R_n^3 , \\ D(\vec{k}) &= (\tfrac{1}{2}d)\sum_n{}' (3\cos^2\theta_n - 1)e^{i\vec{k}\cdot\vec{R}_n}/R_n^3 , \\ E(\vec{k}) &= -\tfrac{3}{2}d\sum_n{}' \sin^2\theta_n e^{-2i\varphi_n} e^{i\vec{k}\cdot\vec{R}_n}/R_n^3 . \end{aligned} \tag{A2}$$

In Eqs. (A2), $d = g^2\mu_B^2 S$. $\sum_n$ and $\sum_n'$ denote summations on lattice points in the same and opposite sublattices, respectively. The polar angles θ_n and φ_n are taken relative to the direction of magnetization. The renormalization terms of Eq. (2) are omitted for the sake of this discussion.

In Eq. (A1) only terms which are first order in $(J_sS)^{-1}$ have been retained, corresponding to the neglect of $\alpha^2 \approx 10^{-5}$ in Eq. (2). Allowing for correction of minor printing errors, Eq. (A1) agrees

with the appropriate specialization of Ziman's result. It reduces to Oguchi's result[40] in his approximation that B and E [Eq. (A2)] are real and yields the dispersion relation given by Harris[20] using his long-wavelength evaluation of A-E. In terms of the lattice sums and combinations defined in Table II, Eq. (A1) may be rewritten for $\vec{k}$ in the (x, y) plane

$$E_{\vec{k}}^{\pm}/4|J_s|S=\{1-\gamma_{\vec{k}}^2+(g\mu_B/2|J_s|S)\times[H_A+(3d/2g\mu_B)(\Sigma_4^{(0)}-\Sigma_1^{(0)})]+(d/4|J_s|S)\Sigma'\pm(3d/4|J_s|S)\Sigma''\}^{1/2}\ . \quad \text{(A3)}$$

In addition to the anisotropy field contribution in the square bracket, we find a slight modification of the form of $E_{\vec{k}}$ as well as a splitting of the two spin-wave branches as expressed by the Σ' and Σ'' terms in Eq. (A3), respectively. The splitting vanishes, however, as $\vec{k}\to\vec{0}$; thus, there is no degenerate spin-wave manifold as is found in the 3D case. Note that Eq. (A3) does not include any effect of zero-point spin reduction on dipolar anisotropy.

The dipolar sums in Table I have been evaluated at a selection of $\vec{k}$ values for the QL structure, with the results presented in Table III. We apply these results to the spin-wave dispersion in K_2MnF_4. Taking $a=5.871$ Å,[1] we find for the dipolar anisotropy field $H_{A(D)}=(\frac{3}{2}g\mu_BS)(\Sigma_4^{(0)}-\Sigma_1^{(0)})=2.57$ kG at $T=0$ K, in good agreement with the calculated value given by Breed.[1] Further, the results of Table III are used to compare $E_{\vec{k}}^{\pm}$ [Eq. (A3) with $H_A=0$] with the simple spin-wave dispersion

$$E_{\vec{k}0}=4|J_s|S[1-\gamma_{\vec{k}}^2+g\mu_BH_{A(D)}/2|J_s|S]^{1/2}\ ,$$

i.e., considering dipolar anisotropy as a single-ion anisotropy. We take $J_s/k_B=-8.7$ K.

Calculated values of $E_{\vec{k}}^{\pm}/E_{\vec{k}0}$ along with the parameters Σ' and Σ'' [Eq. (A3)] are listed in Table IV for the wave vectors given in Table III. The shift in the dispersion is seen to be of the order of 0.1% or smaller at the points sampled and to be roughly independent of polar angle. The mode splitting is seen to be considerably larger in the [11] direction than in the [10], and ranges up to $\approx 0.5\%$ in value.

TABLE II. Dipolar lattice sums in the quadratic layer. Subscripts 1, 2, and 3 refer to sums over lattice points on the same sublattice (excluding the origin); subscripts 4, 5, and 6 refer to sums over the opposite sublattice. Superscripts (0) denote $k=0$.

$$\Sigma_{1,4}=\sum_n[(R_n^2-3z_n^2)/R_n^5]\cos k_xx_n\cos k_yy_n$$

$$\Sigma_{2,5}=\sum_n[(x_n^2-y_n^2)/R_n^5]\cos k_xx_n\cos k_yy_n$$

$$\Sigma_{3,6}=-\sum_n(x_ny_n/R_n^5)\sin k_xx_n\sin k_yy_n$$

$$\Sigma'=\Sigma_4-\Sigma_4^{(0)}-\Sigma_1+\Sigma_1^{(0)}$$

$$\Sigma''=[(\Sigma_5-\gamma_{\vec{k}}\Sigma_2)^2+4(\Sigma_6-\gamma_{\vec{k}}\Sigma_3)^2]^{1/2}$$

TABLE III. Dipolar ($\vec{k}$-dependent) lattice sums for QL structure in units a^{-3}, summed out to $R_n=20a$, where a is the magnetic lattice constant.

$\vec{k}$	k_xa/π	k_ya/π	Σ_1	Σ_2	Σ_3	Σ_4	Σ_5	Σ_6
0	0	0	8.72	0	0	16.20	0	0
$\vec{k}_1$	0.10	0	7.14	−0.65	0	14.57	−0.59	0
$\vec{k}_2$	0.20	0	5.47	−1.23	0	12.72	−1.00	0
$\vec{k}_3$	0.40	0	2.68	−2.25	0	9.24	−1.37	0
$\vec{k}_4$	0.60	0	0.68	−3.04	0	6.00	−1.23	0
$\vec{k}_5$	0.80	0	−0.53	−3.54	0	2.95	−0.71	0
$\vec{k}_6$	1.00	0	−0.94	−3.71	0	0	0	0
$\vec{k}_7$	0.10	0.10	6.44	0	−0.39	13.81	0	−0.50
$\vec{k}_8$	0.20	0.20	4.21	0	−0.64	11.26	0	−1.09
$\vec{k}_9$	0.40	0.40	0.86	0	−0.75	6.73	0	−2.43
$\vec{k}_{10}$	0.60	0.60	−1.23	0	−0.50	3.15	0	−3.80
$\vec{k}_{11}$	0.80	0.80	−2.31	0	−0.16	0.81	0	−4.85
$\vec{k}_{12}$	1.00	1.00	−2.64	0	0	0	0	−5.25

APPENDIX B: RESIDUAL EFFECTS OF DISPERSION ALONG THE AXIS

We analyze the effect on sublattice magnetization behavior of a c-axis exchange coupling J_c between corresponding moments in second-neighbor quadratic layers in the approximation $|J_c/J_s|\ll 1$. The actual form of interlayer coupling is undoubtedly very complex, since many exchange paths are possible. It is expected, however, that our simple model will exhibit the qualitative features to be expected from interlayer coupling.

Following the analysis given by Keffer,[15] the magnon dispersion relation is given by

$$E_{\vec{k}}=[(A_{\vec{k}}-|B_{\vec{k}}|)(A_{\vec{k}}+|B_{\vec{k}}|)]^{1/2}, \quad \text{(B1)}$$

where

$$A_{\vec{k}}=g\mu_BH_A+\tfrac{1}{2}(J_0^{aa}-J_{\vec{k}}^{aa}+J_0^{bb}-J_{\vec{k}}^{bb})-J_0^{ab} \quad \text{(B2)}$$

and

$$B_{\vec{k}}=-J_{\vec{k}}^{ab}\ , \quad \text{(B3)}$$

where $J_{\vec{k}}^{aa}=S\sum_m J_{1m}e^{i\vec{k}\cdot\vec{R}_{1m}}$, etc. The superscripts indicate that summations are to be taken over sites on the same or opposite sublattice in an obvious notation and $\vec{k}$ is here a variable in the 3D reciprocal space rather than in 2D as in the rest of the paper.

We distinguish two cases, one in which J_c is positive (ferromagnetic) in which it is reasonable to assume the c-axis spin pairs are parallel and another with J_c negative, assuming antiferromagnetic spin pair alignment. It is convenient to express the results in terms of exchange fields $H_E=4|J_s|S/g\mu_B$ and $H_{EC}=2|J_c|S/g\mu_B$. We find, then, for the two cases,

$$E_{\vec{k}}=g\mu_B\{[H_A+(1+\gamma_c)H_{EC}+(1+\gamma)H_E]$$

$$\times[H_A+(1-\gamma_c)H_{EC}+(1-\gamma)H_E]\}^{1/2}\ , \quad J_c<0 \qquad \text{(B4)}$$

and

$$E_{\vec{k}}=g\mu_B\{[H_A+(1-\gamma_c)H_{EC}+(1+\gamma)H_E]$$

$$\times[H_A+(1-\gamma_c)H_{EC}+(1-\gamma)H_E]\}^{1/2}, \quad J_c>0, \qquad \text{(B5)}$$

where $\gamma_c=\cos(\frac{1}{2}k_z c)$, $\frac{1}{2}c$ being the separation of second-neighbor planes. First we note that at $k=0$ ($\gamma=\gamma_c=1$), E_0 is modified by an amount $\approx \frac{1}{2}H_{EC}/H_E$ for $J_C<0$ and is unmodified for $J_c>0$. Hence, there are only minuscule effects on the uniform precession mode. The primary case of interest is that of arbitrary k_z with $(k_x^2+k_y^2)^{1/2}$ small, since it is this region of $\vec{k}$ space that controls the low-temperature properties. Thus, we keep only first-order terms in the corrections, putting $\gamma=1$ for them. Equations (B4) and (B5) then *both* reduce to

$$E_{\vec{k}}/4|J_s|S\approx\{1-\gamma^2+2[H_A+(1-\gamma_c)H_{EC}]/H_E\}^{1/2}, \qquad \text{(B6)}$$

and we find as expected a modified anisotropy field, which, however, is k_z dependent. Note that anisotropy is *increased* regardless of the sign of J_c.

The major conclusion to be drawn here is that thermodynamic measurements, which presumably average over-all allowed values of k_z, will yield a spin-wave gap energy different from AFMR values, i.e., larger by an amount $\approx |J_c|/g\mu_B H_A$. To see this in some detail we recall the approximation Eq. (14) to the low-temperature deviation in sublattice magnetization. This result can be obtained with $E_{\vec{k}}$ from Eq. (B6) leaving the integration over k_z to the end, giving for both $J_c>0$ and $J_c<0$ the result

$$\Delta S(T)\approx -\frac{(1+\alpha)k_B T}{\pi^2|J_s|S}\int_0^{\pi/2} d\theta$$

$$\times\ln\{1-\exp[-T_G(1+2H_{EC}\sin^2\theta/H_A)^{1/2}/T]\}\ . \qquad \text{(B7)}$$

To first order in H_{EC}/H_A, $\sin^2\theta$ in Eq. (B7) may be replaced by its average value. Equation (B7) then returns to the form of Eq. (14) with $T_G \to T_G(1+H_{EC}/2H_A)$. This "effective energy gap" approximation is very good for small H_{EC}. Discussion of possible c-axis coupling in the QL compounds is found in Secs. IV and V.

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Antiferromagnetic Resonance in the Quadratic-Layer Antiferromagnets K_2MnF_4 and Rb_2MnF_4

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Antiferromagnetic resonance has been observed in the quadratic-layer antiferromagnets K_2MnF_4 ($T_N = 42.1$ K) and Rb_2MnF_4 ($T_N = 38.4$ K) at a frequency of 24 GHz as a function of the temperature from 1.5 up to 38 and 34 K, respectively, with the static magnetic field parallel to the c axis. To reduce these data to zero-field magnon energy gaps, the frequency renormalization method of Oguchi including a static magnetic field was used. The resulting equations in frequencies and spin-wave occupation numbers were solved self-consistently. The gaps as a function of the temperature appear to scale approximately with the magnetization. By extrapolation, the gaps at $T = 0$ K were found to be 7.40 ± 0.05 K for K_2MnF_4, and 7.28 ± 0.05 K for Rb_2MnF_4. The antiferromagnetic-resonance linewidths increase steadily in approaching T_N.

I. INTRODUCTION

Antiferromagnetism in two dimensions has received a great deal of attention in the last few years. The best examples to date of nearly Heisenberg two-dimensional antiferromagnetism appear to be the family of the quadratic-layer (QL) structures, the principal members of which are K_2NiF_4, K_2MnF_4, and Rb_2MnF_4. Neutron-diffraction studies have shown that there is a negligible dispersion of spin waves in these systems along the tetragonal axis,[1] while precision measurements of the temperature dependence of the sublattice magnetization[2] show that two-dimensional spin-wave theory is applicable at least in the lower half of the ordered region. Further, it has been established recently by a double-resonance experiment,[3] that the zero-point spin reduction is in accord with the spin-wave result for the quadratic layer.

An important quantity in spin-wave theory is the $k = 0$ magnon energy, which usually enters into the analysis of a thermodynamic quantity in terms of spin-wave theory as an independent variable. It is the purpose of the present paper[4] to determine experimentally the magnon energy gap, and its temperature dependence, for K_2MnF_4 ($T_N = 42.1$ K) and Rb_2MnF_4 ($T_N = 38.4$ K), compounds in which the anisotropy results dominantly from magnetic dipole-dipole interactions ($H_A \approx 2700$ G). Unfortunately, a direct precision measurement of the gap in zero external magnetic field is not readily feasible ($E_{gap}/k_B \approx 7$ K).[5] Instead, we have observed antiferromagnetic resonance (AFMR) of the lower spin-wave branch close to the spin-flop field (≈ 55 kG) in a microwave experiment ($h\nu/k \approx 1.4$ K), and derived the zero-field gaps by use of spin-wave theory. To carry out this reduction the frequency renormalization method of Oguchi[6] was first generalized to include a static magnetic field, and the resulting equations were then solved self-consistently. Finally, data on the AFMR linewidth are presented.

II. EXPERIMENTAL

The experiments were carried out with a superheterodyne EPR spectrometer of standard design, operating at 24 GHz and equipped with phase-sensitive detection of the intermediate frequency. The spectrometer was tuned to the absorption mode.