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OBSERVATION OF ZERO-POINT SPIN REDUCTION IN QUADRATIC LAYER ANTIFERROMAGNETS

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Nuclear magnetic resonance of ^{55}Mn nuclei, detected through its effect on the antiferromagnetic resonance, has been used to measure the zero-point spin reduction in the quadratic layer antiferromagnets K_2MnF_4 and Rb_2MnF_4. The result, including a small correction for supertransferred hyperfine effects, is $S-\langle S_z\rangle_{T=0}=0.17\pm0.03$, in excellent agreement with spin-wave theory.

In this Letter we report measurements of the zero-point spin reduction in the quadratic layer antiferromagnets K_2MnF_4 and Rb_2MnF_4, obtained through the NMR of the ^{55}Mn nuclei. Recent studies[1,2] of these compounds, and their isomorph K_2NiF_4, have shown their magnetic properties to be exclusively two-dimensional. Their magnetic structure being well defined and spin-wave (as well as perturbation) theory being readily applicable, they are excellent cases for comparing theoretical predictions of zero-point spin reduction with experiment. In addition, the reduction is expected to be larger for the quadratic layer structure than for the three-dimensional structures,[3] whereas supertransferred hyperfine interactions,[4] which tend to mask the effect, are smaller. The present measurements are the first which allow an unambiguous comparison with theory, and yield a value for $\Delta_0=S-\langle S_z\rangle_{T=0}$ that, including a small correction for supertransferred hyperfine effects, agrees well with spin-wave theory.

The method used here to probe $\langle S_z\rangle$ is to measure the NMR frequency of the ^{55}Mn nuclei, which—in zero field, and apart from a number of small corrections to be discussed later—is given by $\omega_n=\gamma_n H_n=|A\langle S_z\rangle/\hbar|$, where the hyperfine constant A is calibrated in a magnetically diluted isomorph. The NMR has been observed[5] by the electron-nuclear double-resonance (ENDOR) method first employed by Heeger et al.[6] in studying $KMnF_3$. The method relies on the contribution $H_{An}=A\langle I_z\rangle/\gamma_e\hbar$ to the anisotropy field H_A made by the ^{55}Mn nuclei via the hyperfine interaction $A\vec{S}\cdot\vec{I}$, i.e., $H_A=H_{Ae}+\frac{1}{2}(H_{An+}+H_{An-})$, where $H_{An\pm}$ are the anisotropy fields due to the nuclei at thermal equilibrium. Here, (+) and (−) refer to the nuclei on the sublattices with electron spins parallel and antiparallel, respectively, with the external field applied along the c axis. H_{Ae} is the anisotropy field from other sources, mainly arising from electronic dipolar interactions ($H_{Ae}\approx 2.3$ kG).[7] The external field H_0 required for antiferromagnetic resonance (AFMR), given by $\Omega_{1,2}=\Omega_{gap}\pm\gamma_e H_0$ with $\Omega_{gap}=\gamma_e(2H_E H_A+H_A^2)^{1/2}$, can therefore be shifted by saturating the ^{55}Mn nuclei with a resonant rf field, thus making possible the detection of the ^{55}Mn NMR. The shift is given by

$$|\Delta H_0|=\gamma_e(H_E+H_A)\times(H_{An+}\eta_+ + H_{An-}\eta_-)/2\Omega_{gap}, \qquad (1)$$

where $\eta_\pm$ is the fractional decrease of the nuclear polarization due to the applied rf power. The exchange field H_E has been derived from Breed's[7] exchange coupling constants with proper care for renormalization; Ω_{gap} has been taken from recent AFMR experiments.[8] In the present case, where $H_{An\pm}T\approx 9$G °K, there results $|\Delta H_0|\approx 30$ G at 1.5°K for full saturation of the (+) or (−) nuclei.

Single crystals of K_2MnF_4 and Rb_2MnF_4 were mounted in a microwave cavity resonating at 23.26 GHz with the c axis oriented along the applied field to within 1°. The experiments were

Table I. Experimental NMR frequencies and linewidths at the field values quoted, nuclear spin-wave shifts, and pure hyperfine frequencies. All entries are in megahertz. Temperature is 1.45°K.

$\nu_\pm(0)$[a]	$\Delta\nu_\pm(0)$[a,b]	$[\nu_\pm(1)-\nu_\pm(0)]$ (calc)	$[\nu_\pm(1)-\nu_\pm(0)]$ (meas)	$\nu_\pm(1)$	ν_n
		K_2MnF_4 ($H_0=47.1$ kG)			
687.6 ± 0.3	4.7	3.4	3.2 ± 0.6	691.0	642.5
591.5 ± 0.3	4.7	2.4	2.6 ± 0.5	593.9	
		Rb_2MnF_4 ($H_0=46.1$ kG)			
685.7 ± 0.3	5.5	2.9	3.0 ± 0.6	688.6	641.2
591.8 ± 0.3	5.4	2.0	2.2 ± 0.4	593.8	

[a] Actually measured at $\eta_\pm \approx 0.02$ at the peak.
[b] Full width at half-maximum.

carried out at 1.45°K with $H_0 \approx 47$ kG required to bring the low-lying $k=0$ spin-wave branch (Ω_2) to resonance with the microwave radiation. The field was set at the maximum slope of the AFMR line (linewidth ≈800 G) to permit its shift to be recorded as an amplitude variation. A standard ENDOR arrangement was used to apply rf power (H_1 up to ≈2 G) to the samples at the NMR frequencies. The results (Table I) were unaffected by small changes (≈1°) in crystal orientation.

The electron-nucleus interaction produces an indirect coupling among the nuclei ("nuclear spin waves") through the electron spins, which leads to a displacement of the NMR lines proportional to the nuclear polarization. Although the effect is quite small in the present case, it is necessary to correct for it in order to determine the pure hyperfine frequency ω_n. The effect, observed previously in other systems,[9] was first calculated by de Gennes et al.[10] The "pulled" frequencies of the two NMR modes are

$$\omega_\pm(\eta_\pm)=\gamma_n H_n[1-\gamma_e^2(H_E+H_A)H_{An\pm}(1-\eta_\pm)/\Omega_1\Omega_2 P_\pm]\pm\gamma_n H_0[1-\gamma_e^2 H_n H_{An\pm}(1-\eta_\pm)/\Omega_1\Omega_2 P_\pm], \quad (2)$$

where $P_\pm=1-2\gamma_e\gamma_n H_0(H_0\pm H_n)/\Omega_1\Omega_2$ is a factor very close to unity. All parameters in Eq. (2) are defined to be positive. Note that the two sublattices are decoupled as far as their effects on the NMR modes are concerned. Nuclear spin-wave shifts upon full saturation ($\eta_\pm=1$), calculated from Eq. (2), have been entered in Table I, and applied to the experimental frequencies at vanishing rf power $\nu_\pm(0)=\omega_\pm(0)/2\pi$ to obtain the "unpulled" frequencies $\nu_\pm(1)$ and their averages $\frac{1}{2}[\nu_+(1)+\nu_-(1)]=\nu_n=\omega_n/2\pi$, from which the spin deviation is to be derived.[11]

The pulling effect has also been determined experimentally in the following way. The AFMR line shift is measured versus the NMR frequency at a set of fixed power levels. Since both the AFMR shift, Eq. (1), and the pulling effect, Eq. (2), are linear in $\eta_\pm$, a set of tilted curves is obtained, as is shown in Fig. 1 for the high-frequency NMR branch of Rb_2MnF_4. The peaks of these curves obviously lie on a straight line intersecting the abscissa at $\nu_\pm(0)$. To establish the limit of full saturation $\nu_\pm(1)$, we rely on the fact that at the peaks $\eta=\sigma/(1+\sigma)$ with σ proportional to the rf power. This procedure has been verified using absolute measurements of the AFMR shift and Eq. (1). Nuclear spin-wave shifts, thus measured, are in good agreement with the calculated ones (Table I). At low power levels ($\eta \ll 1$) the measured ΔH_0 simply plot out the unbroadened NMR absorption line shape, and from this the NMR linewidth has been measured (Table I).[12]

In order to deduce $\langle S_z\rangle_{T=0}$, the ν_n values quoted in Table I are extrapolated to zero temperature by use of the low-temperature analytical expression for the sublattice magnetization derived earlier,[2] suitably modified to include effects of H_0 on the spin-wave spectrum. After this minor correction,[13] we have $\nu_n(T=0)=643.5\pm1.0$ MHz for K_2MnF_4 and 642.4 ± 1.0 MHz for Rb_2MnF_4. The hyperfine structure constant A appropriate to these compounds may be expressed as $A=A_{loc}+A_{dip}+A_{sthf}$, where A_{loc} represents the single-ion hyperfine interaction, and A_{dip}

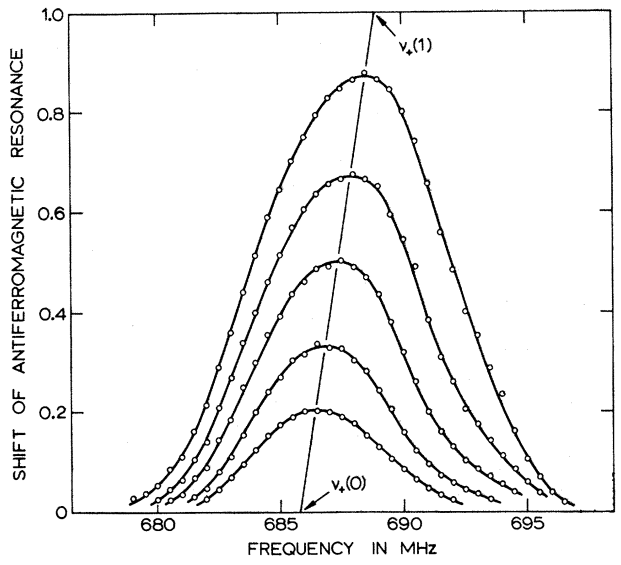

FIG. 1. The shift of the AFMR line (towards lower fields) versus the frequency of the rf power for a series of different power levels, upon saturation of the high-frequency NMR branch (ω_+) in Rb_2MnF_4. The vertical scale, in relative units, corresponds to η_+, the fractional decrease of the polarization of the ^{55}Mn nuclei. The straight line, drawn through the peaks of the curves, represents the shift of the NMR line with η_+.

and A_{sthf} result from dipolar and supertransferred hyperfine fields, respectively, due to neighboring Mn ions. A_{loc} can be closely estimated from recent ENDOR studies of Mn^{2+} diluted in $KMgF_3$ and K_2MgF_4,[14] which yield $A = -273.85 \pm 0.02$ MHz and -272.24 ± 0.05 MHz, respectively, at 4.2°K. From these values we adopt $A_{loc} = -273.0$ MHz $\pm 1\%$ for the present compounds. Systematic observation of Mn^{2+} in sixfold fluorine coordination[15] indicates that A is very much independent of the host lattice, with a slight trend of increasing $|A|$ with dilation of the fluorine environment. Since the Mn-F distances in K_2MnF_4 and Rb_2MnF_4 are ≈5% greater than those in K_2MgF_4, $|A_{loc}|$ might be slightly larger than, but within the quoted uncertainty of, the adopted value. This would, of course, correspondingly increase the spin deviation deduced. The dipolar contribution to A is calculated by lattice summation, giving $A_{dip} = +0.72$ MHz for K_2MnF_4 and $+0.69$ MHz for Rb_2MnF_4. The supertransferred hyperfine coupling is estimated from the calculations of Huang et al.[16] for $KMnF_3$, noting that the Mn-F-Mn distances and exchange couplings[7,17] are very nearly the same as those in K_2MnF_4. Scaling by a factor $\frac{2}{3}$ to the fourfold Mn coordination of the quadratic layer, we find $A_{sthf} = -3.2$ MHz. This is only 1% of A_{loc}, and could be subject to a substantial error without seriously affecting our results. Summarizing, we have $A = -275.5$ MHz $\pm 1\%$, and obtain, for both compounds,

$$\Delta_0 = 0.17 \pm 0.03.$$

It is noted that the error mainly resides in the uncertainty of the hyperfine coupling.

The final number is in excellent agreement with the value $\Delta_0 = 0.17$ obtained from spin-wave theory for the values of $\alpha = H_A/H_E$ appropriate to the compounds studied here.[18] It is significantly larger, however, than the result found with perturbation theory by Davis[19] and Walker[20] for the quadratic layer with $S = \frac{5}{2}$, which, including terms up to the sixth order, is $\Delta_0 \approx 0.11$. Since these calculations are formally exact when evaluated to all orders, the higher-order terms, which are impractical to evaluate, are apparently of considerable importance in calculating Δ_0.

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[12]The measured linewidth is in general accord with Suhl-Nakamura interaction (L. R. Walker, unpublished). Quadrupolar splitting is expected to be small [A. H. M. Schrama, P. I. J. Wouters, and H. W. de Wijn, Phys. Rev. B 2, 1235 (1970)].

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GROUND-STATE BANDS IN NEUTRON-RICH EVEN Te, Xe, Ba, Ce, Nd, AND Sm ISOTOPES PRODUCED IN THE FISSION OF ^{252}Cf†

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We present experimental results on the ground-state bands of heavy even-even nuclei produced in the primary fission of ^{252}Cf. Experimental values for the energy levels and lifetimes range from those typical of spherical nuclei to those associated with permanently deformed nuclei.

In this Letter we present new information concerning the energy levels of very neutron-rich even-even isotopes of ^{52}Te, ^{54}Xe, ^{56}Ba, ^{58}Ce, ^{60}Nd, and ^{62}Sm. These results were obtained in a series of experiments on the prompt gamma-ray de-excitation of the fission fragments from spontaneous fission of ^{252}Cf. The data, which in some of the cases can be correlated with previously reported results, extend the knowledge about the systematic behavior of collective excitations to neutron-rich nuclei far from the β stability line. The systematics of the energy levels in the ground-state bands for the heavier fragments are well fitted using the phenomenological variable-moment-of-inertia model of Mariscotti, Scharff-Goldhaber, and Buck.[1] One of the main features of the results is the evidence that the well-known abrupt discontinuity in the ratio $E4^+/E2^+$ for isotopes with 88 and 90 neutrons reaches its maximum effect in Nd, Sm, and Gd isotopes and becomes much less abrupt in the Ce and Ba nuclei. This smoother transition is similar to the behavior observed for isotopes with $Z > 66$.[2]

In the experiments x rays and/or γ rays were measured in coincidence with pairs of fission fragments. The experimental technique has been described in a previous paper[3] and will therefore only be briefly summarized here. In most of the cases the atomic number was determined by observing a coincidence between the characteristic K x rays and one or more of the γ rays of the ground-state band. The masses of the fragments were calculated from their measured kinetic energies. Direct determination of lifetimes in the region of 0.2-2 nsec were obtained from Doppler-shift considerations.

The experimental results are presented in Table I. For each isotope in the table we present two lines of information. The top line contains the experimental energies of the observed levels along with the ratio of the energies of the 4^+ and 2^+ levels, the measured half-life of the 2^+ level, the yield per fission of this transition (corrected for internal conversion), and the mean experimental mass associated with the ground-state-band transitions. Also presented are $B(E2; 2 \rightarrow 0)$ values. The second line contains corresponding predicted values. The energies of the