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Preprint

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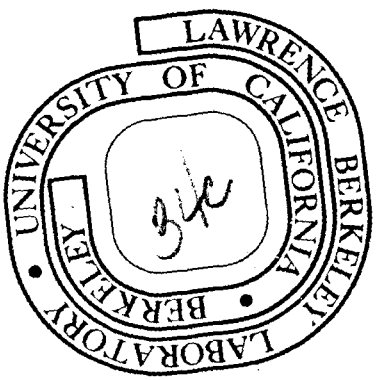

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DISCLAIMER

PARAMAGNETIC SHIFTS AND SPIN-FLOP IN SUPERTRANSFERRED HYPERFINE STRUCTURE OF ^{111m}Cd in $RbMnF_3$*

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The signs of the supertransferred hyperfine fields at the Cd nucleus in paramagnetic $RbMnF_3$, $KFeF_3$, and $KCoF_3$ have been measured. In the antiferromagnetic phase of $RbMnF_3$ the effect of the spin-flop transition on the PAC of ^{111m}Cd has been observed.

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Recently [1] we reported the PAC of ^{111m}Cd doped as a substitutional impurity into the antiferromagnetic perovskites $RbMnF_3$, $KCoF_3$ and $KNiF_3$. The hyperfine field at the Cd nucleus is caused by unpaired spin density in outer (4s,5s) Cd s-orbitals, transferred from the six nearest magnetic neighbors along linear $Mn^{2+} - F^- - Cd^{2+}$ bonds. Since the contact field produced by an s electron is opposite to its spin, the hyperfine field at the Cd nucleus should be parallel to the magnetic moment of the six nearest transition metal ions. In the antiferromagnetic state only the magnitude of the supertransferred hyperfine interaction is obtained, but the sign can be measured in the paramagnetic state by applying an external magnetic field.

In this Letter we report the time differential PAC of ^{111m}Cd doped into paramagnetic, polycrystalline $RbMnF_3$. The spectra (Fig. 1) were observed at 300°K and 87°K (close to the Néel point T_N = 83°K), with an external field H_{ext} = 31.3 kOe applied perpendicular to the detector axis. In fig. 1 the perturbation factor [2], $A_{22}G_{22}(t) = A_{22}(0.25 + 0.75\cos(4\pi\nu_L t)$ is shown for ^{111m}Cd in $RbMnF_3$ and for a diamagnetic standard ($CdCl_2$ solution) in the same external field. The higher frequency of ^{111m}Cd in $RbMnF_3$ corresponds to a higher effective field $H_{eff} = H_{ext} + \Delta H$. This paramagnetic shift arises from a supertransferred contact field at the Cd nucleus created by polarization of the Mn^{2+} electron spin S = 5/2. Since the electronic spin fluctuations are fast compared to the characteristic time of the PAC experiment, only the effect of the time average $\langle S_z \rangle$ is seen. Therefore, $\Delta H = H_{hf}(4°K)\ \langle S_z \rangle/2.5$, where $H_{hf}(4°k)$ = 116 kOe is taken as the low-temperature limit of the hyperfine field at the Cd nucleus in the antiferromagnetic state and $\langle S_z \rangle$ = 5/2 has been used as the local value of $\langle S_z \rangle$ for T = 4°K. (A small zero-point spin deviation has been neglected in this estimate). The spin expectation value $\langle S_z \rangle = (-\chi_m H_{ext})/(g\beta N_L)$ in the paramagnetic state can be estimated using the molar susceptibility $\chi_m = C/(T+\theta)$. In this way we estimate for ^{111m}Cd in $RbMnF_3$, at 87°K, ΔH_{est} = +2.0 kOe compared to ΔH_{obs} = +3.0±0.5 kOe. The lower susceptibility at room temperature corresponds to a smaller paramagnetic shift (Fig. 1). In this way the following relative shifts $\Delta H/H_{ext}$ were observed; $RbMnF_3$: +9.6±1.6% (87°K), +5.1±1.6% (300°K); $KFeF_3$: +5.4±2.2% (120°K); $KCoF_3$: +4.5±1.6% (120°K) and

$KNiF_3$: +1.3±1.6% (265°K). Because of the lower susceptibilities, smaller paramagnetic shifts were observed in $KFeF_3$, $KCoF_3$ and $KNiF_3$ compared to $RbMnF_3$/Cd. For $KNiF_3$ the large values of θ and T_N led to a paramagnetic shift that lay within our experimental error. The experiments reported here are analogous to paramagnetic shift measurements that have been made on F nuclei in these lattices, using NMR. Because the Cd field arises from supertransferred hyperfine structure, these shifts provide a stringent test of the proposed mechanism [1] for spin transfer through $Mn^{2+} - F^- - Cd^{2+}$ bonds.

$RbMnF_3$ is a well known antiferromagnet with an exceptionally low critical field H_c = 2.45 kOe for the spin-flop transition. Above this value the spin axis lies perpendicular to the external field. In polycrystalline $RbMnF_3$/Cd (4°K) the hyperfine fields at the Cd are randomly oriented for $H_{ext} = 0$, whereas they are confined in a plane perpendicular to the external field for $H_{ext} > H_c$. Since PAC is sensitive to the orientation of the hyperfine fields relative to the detector axis, this method lends itself to the observation of the spin-flop transition, as shown (Fig. 2). The time spectra were taken with the detectors at 180° parallel to H_{ext}. In this case the perturbation factor can be written as $A_{22}G_{22}(t) = A_{22}(1-b_1-b_2 + b_1\cos(2\pi\nu_L t) + b_2\cos(4\pi\nu_L t))$. The Fourier coefficients b_1, b_2 are equal for the random distribution $b_1 = b_2 = 0.4$ ($H_{ext} = 0$) and $b_1 = 0$, $b_2 = 0.75$ if the hyperfine fields are in a plane perpendicular to H_{ext}. The spin-flop transition was observed as a change in the ratio b_1/b_2 of the Fourier coeffi-

cients (Fig. 2). Since a polycrystalline sample was used, the transition is fairly smeared out; for a precise determination of H_c a single crystal should be used.

This observation gives further microscopic support of both the proposed spin-transfer mechanism and the accepted model for spin-flop transitions.

References

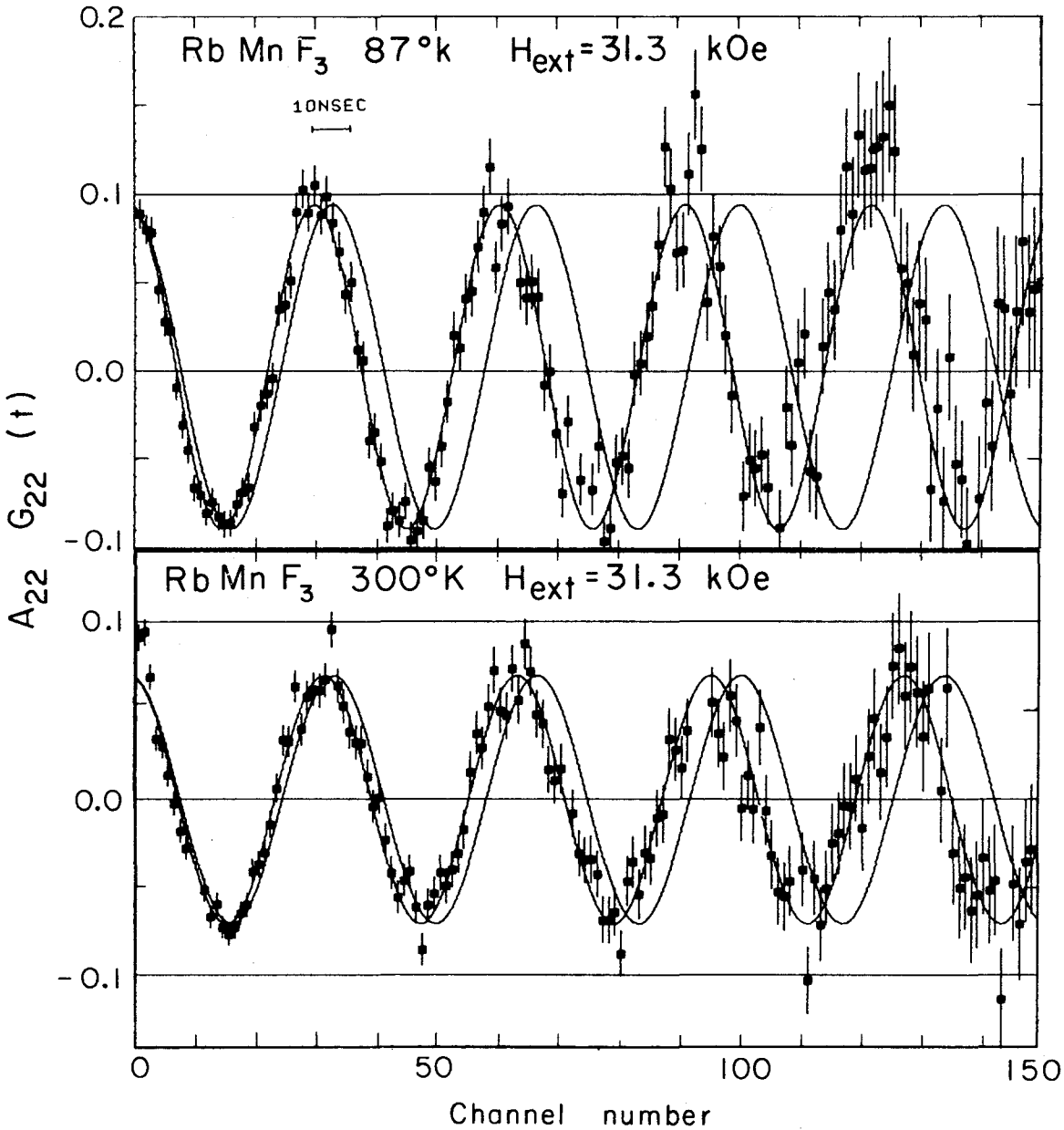
1. H. H. Rinneberg and D. A. Shirley, Phys. Rev. Letters 30 (1973) 1147.

2. H. Frauenfelder and R. M. Steffen, In Alpha-, Beta-, and Gamma-Ray Spectroscopy, ed. by K. Siegbahn (North-Holland, 1965), Vol. 2.

Figure Captions

Fig. 1. PAC of ^{111m}Cd in paramagnetic $RbMnF_3$, compared to a diamagnetic standard ($CdCl_2$ solution) in the same external field H_{ext} = 31.3 kOe.

Fig. 2. Spin-flop transition in $RbMnF_3$ detected by PAC of ^{111m}Cd.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10NSEC
Rb Mn F3 300°K Hext=31.3 kOe
A22 G22 (t)
0.2
0.1
0.0
-0.1
0
50
100
150
Channel number
XBL748-3990

Fig. 1

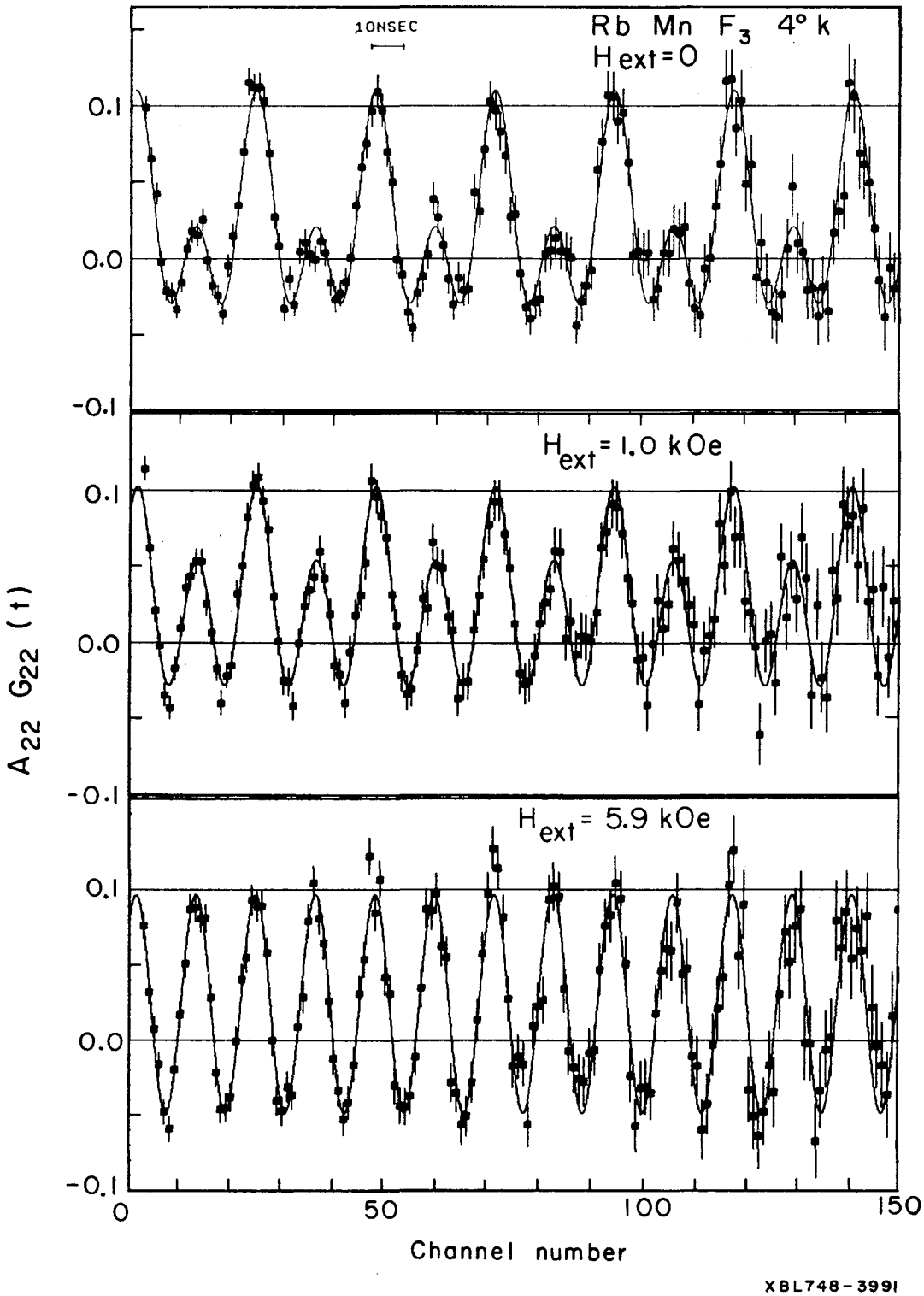
10NSEC
Rb Mn F_3 4° k
Hext=0
H_{ext} = 1.0 kOe
H_{ext} = 5.9 kOe
A_{22} G_{22} (t)
0.1
0.0
-0.1
0
50
100
150
Channel number
XBL748-3991

Fig. 2

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