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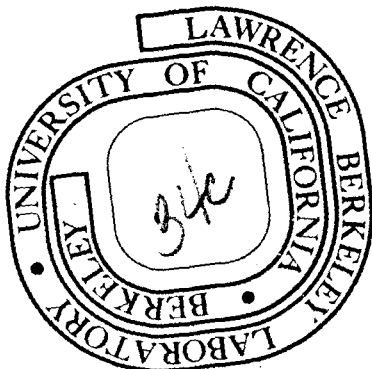
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PARAMAGNETIC SHIFTS AND SPIN-FLOP IN SUPERTRANSFERRED

HYPERFINE STRUCTURE OF ^{111}mCd 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 ^{111}mCd has been observed.

Recently [1] we reported the PAC of ^{111}mCd 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 $\text{Mn}^{2+} - \text{F}^- - \text{Cd}^{2+}$ 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 $T_N = 83^\circ\text{K}$), with an external field $H_{\text{ext}} = 31.3\text{ kOe}$ 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 $H_{\text{eff}} = H_{\text{ext}} + \Delta H$. 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, $\Delta H = H_{\text{hf}}(4^\circ\text{K}) \langle S_z \rangle / 2.5$, where $H_{\text{hf}}(4^\circ\text{K}) = 116\text{ kOe}$ 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 $T = 4^\circ\text{K}$. (A small zero-point spin deviation has been neglected in this estimate). The spin expectation value $\langle S_z \rangle = (-\chi_m H_{\text{ext}}) / (g\beta N_L)$ 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 , $\Delta H_{\text{est}} = +2.0\text{ kOe}$ compared to $\Delta H_{\text{obs}} = +3.0 \pm 0.5\text{ kOe}$. The lower susceptibility at room temperature corresponds to a smaller paramagnetic shift (Fig. 1). In this way the following relative shifts $\Delta H / H_{\text{ext}}$ were observed; RbMnF_3 : $+9.6 \pm 1.6\%$ (87°K), $+5.1 \pm 1.6\%$ (300°K); KFeF_3 : $+5.4 \pm 2.2\%$ (120°K); KCoF_3 : $+4.5 \pm 1.6\%$ (120°K) and

KNiF_3 : $+1.3 \pm 1.6\%$ (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 $\text{Mn}^{2+} - \text{F}^- - \text{Cd}^{2+}$ bonds.

RbMnF_3 is a well known antiferromagnet with an exceptionally low critical field $H_C = 2.45 \text{ kOe}$ 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 $H_{\text{ext}} = 0$, whereas they are confined in a plane perpendicular to the external field for $H_{\text{ext}} > H_C$. 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 $b_1 = b_2 = 0.4$ ($H_{\text{ext}} = 0$) 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-

icients (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.

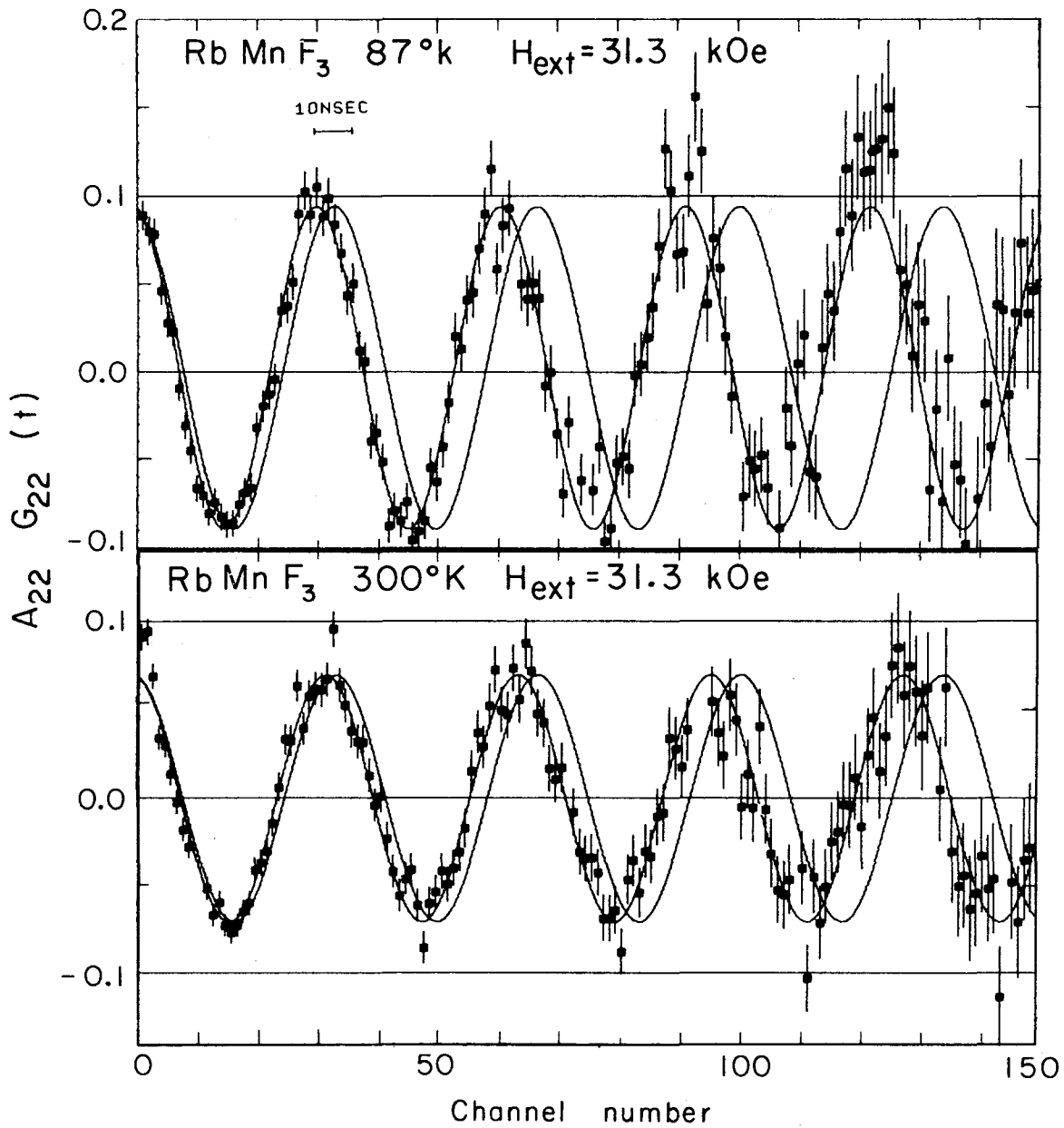
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1. H. H. Rinneberg and D. A. Shirley, Phys. Rev. Letters 30 (1973) 1147.
2. H. Frauenfelder and R. M. Steffen, In Alpha-, Beta-, and Gamma-Ray Spectroscopy, ed. by K. Siegbahn (North-Holland, 1965), Vol. 2.

Figure Captions

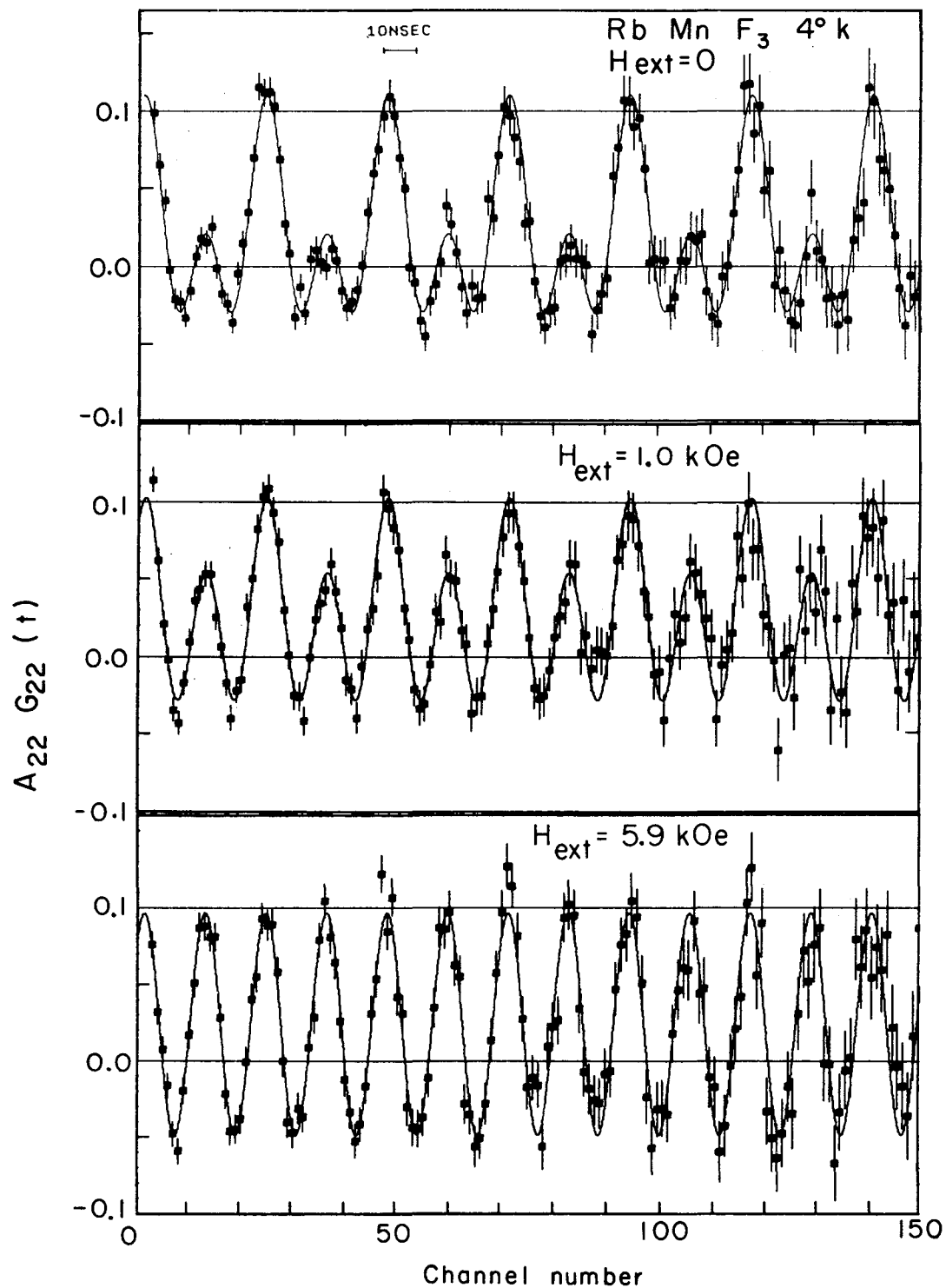
Fig. 1. PAC of ^{111m}Cd in paramagnetic RbMnF_3 , compared to a diamagnetic standard (CdCl_2 solution) in the same external field $H_{\text{ext}} = 31.3 \text{ kOe}$.

Fig. 2. Spin-flop transition in RbMnF_3 detected by PAC of ^{111m}Cd .



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Fig. 1



XBL748-3991

Fig. 2

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