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# MAGNETIC-PHASE DIAGRAM OF $MnF_2$ FROM ULTRASONIC AND DIFFERENTIAL MAGNETIZATION MEASUREMENTS

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The magnetic-phase diagram, in the $H$-$T$ plane, of $MnF_2$ was measured using ultrasonic and differential magnetization techniques. The paramagnetic-antiferromagnetic boundary, for $\vec{H}$ along the $c$ axis, is well represented by $T_N - T = (1.65 \pm 0.15) \times 10^{-10} H^2$ °K/G$^2$. The triple point is at $T_3 = 65.1 \pm 0.2$°K and $H_3 = 120 \pm 4$ kG. The phase boundaries are discussed in light of existing theories.

In this Letter we report on high-field studies of the magnetic-phase boundaries of the classic uniaxial antiferromagnet $MnF_2$ using ultrasonic and differential magnetization techniques. The new results include (1) the observation of an attenuation peak for ultrasonic waves at the paramagnetic transition in finite magnetic fields up to the high-field triple point ($T_3 = 65.1 \pm 0.2$°K, $H_3 = 120 \pm 4$ kG) and (2) the determination of the complete boundaries, in the $H$-$T$ plane, between the paramagnetic and antiferromagnetic phases and between the antiferromagnetic and spin-flop phases. When plotted in a normalized form the paramagnetic-to-antiferromagnetic boundary in $MnF_2$ is shown to be similar to the measured boundaries in other antiferromagnets containing $Mn^{++}$. An analysis of the antiferromagnetic to spin-flop boundary shows that the magnetic-field dependence of the susceptibility should affect this phase boundary near the triple point.

$MnF_2$ has a tetragonal lattice and is antiferromagnetic below the Néel temperature $T_N = 67.4$°K. The anisotropy energy of this material is very small compared with the exchange energy, and it is uniaxial with the $c$ axis (tetragonal axis) as the easy axis for the sublattice magnetizations. For such a material the magnetic-phase diagram in the $H$-$T$ plane, when the applied magnetic field $\vec{H}$ is along the $c$ axis, should consist of three phases[1]: paramagnetic (P), antiferromagnetic (AF), and spin-flop (SF). In the P phase the magnetizations of the two sublattices point along the $c$ axis, are parallel to each other, and have equal magnitudes. In the AF phase the sublattice magnetizations are along the $c$ axis but are antiparallel to each other. In the SF phase, and when $H$ is small compared with the exchange field $H_E$, the sublattice magnetizations are roughly antiparallel to each other and are almost perpendicular to the $c$ axis. The AF-SF transition is a first-order transition which is accompanied by an abrupt change in the magnetic moment.[2] Calculations based on the molecular-field approximation show that the P-AF transition is a second-order transition in the Ehrenfest sense. However, there are experimental data which indicate that this transition is lambdalike both at zero and at finite magnetic fields, at least for some antiferromagnets.[1,3]

Previous experimental investigations of the P-to-AF transition of antiferromagnets were largely confined to water-containing salts with complicated unit cells and with Néel temperatures in the liquid-helium range. We chose $MnF_2$ because it is a classic uniaxial antiferromagnet which has a simple magnetic structure and because many of the magnetic properties of $MnF_2$ had been investigated extensively. Previous investigations of the magnetic phase transitions in $MnF_2$ include (1) the AF-SF transition which was observed magnetically[2] and ultrasonically,[4] (2) the P-AF transition at $T_N$ (in zero field) which was observed by a variety of means including ultrasonic-attenuation measurements,[5] and (3) NMR studies[6] of the P-AF transition in fields up to 8 kG.

The ultrasonic attenuation (UA) of 10- and 30-MHz longitudinal waves propagating along the $c$ axis was measured in liquid nitrogen in steady magnetic fields up to 140 kG. Standard ultrasonic pulse techniques were used. The temperature was measured with a platinum resistance thermometer and corrections for the magnetoresistance were applied when necessary. The uncertainty in the temperature was less than 0.1°K, and the uncertainty in $H$ was less than 1%. Two single crystals of $MnF_2$, obtained from different sources, gave virtually the same results.

A peak in the UA was observed at the P-AF transition both at $H=0$ and in a finite magnetic field applied along the $c$ axis. Going from the P phase to the AF phase, either at constant $H$ by decreasing $T$ or at constant $T$ by decreasing $H$, the UA increases quickly, passes through a sharp maximum at the transition, and then decreases gradually. A trace of the UA as function of $H$ (at constant $T$) is shown in Fig. 1. The type of behavior shown in this figure was observed only at $65°K \lesssim T < T_N$. The position of the attenuation maximum in the $H$-$T$ plane is shown in Fig. 2(a). From these measurements $T_N = 67.43 \pm 0.1°K$.

The AF-SF transition was studied ultrasonically only at 4.2°K and 20.3°K. This transition was accompanied by a sharp peak in the UA, as reported earlier.[4]

The differential magnetic moment, $dM/dH$ (hereafter called DMM), was measured with 10-

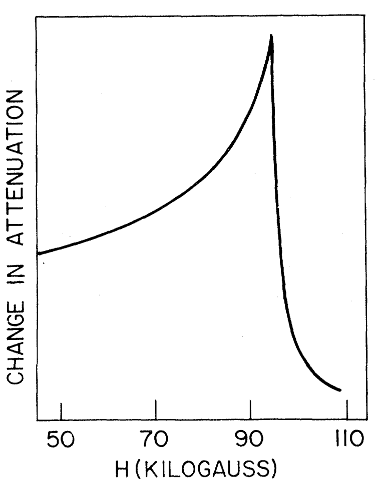


FIG. 1. Recorder tracing of the attenuation of a 30-MHz longitudinal sound wave, in $MnF_2$. The direction of sound propagation and $\vec{H}$ are both parallel to the $c$ axis. The recorder response to the attenuation is nonlinear.

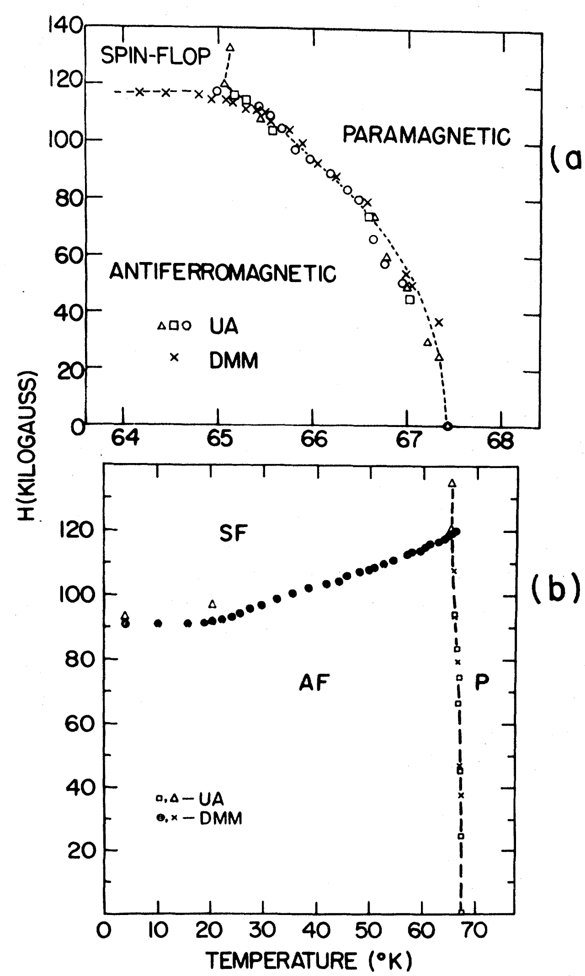


FIG. 2. (a) and (b) Phase boundaries of $MnF_2$ for $\vec{H}$ along the $c$ axis. Part (a) shows an expanded view of the phase boundaries near $T_N$; the dashed line between the P and AF phases corresponds to Eq. (1) with $D = 1.65\times10^{-10}$ °K/G$^2$.

msec half-period pulsed magnetic fields up to 220 kG. The magnetic field, which was along the $c$ axis, was known to within 1%. The susceptibility of $MnF_2$ is sufficiently small so that the demagnetizing fields are expected to be less than ~1%. Data from 64°K up to $T_N$ were taken in liquid nitrogen, and those from 4.2°K up to ~64°K were obtained by first cooling the sample with liquid helium and then letting it warm up slowly (about 2 h from 4.2°K to 70°K). Temperatures were measured with a Cu-Constantan thermocouple.

For temperatures just below $T_N$, the DMM had a step at the P-AF transition. However, as $T$ approached $T_3$ the character of the DMM anomaly at the P-AF transition changed smoothly from a step to a spike. The AF-SF transition was indicated by a sharp peak in the DMM. Attempts to observe the SF-P transition at $T < T_3$ by means of DMM measurements were not successful, i.e., no anomaly in the DMM was observed in pulsed-field scans to 220 kG. The magnetic phase transitions observed by DMM measurements are shown in Fig. 2. These results agree with the UA data.

The P-AF boundary in the $H$-$T$ plane is well represented by the relation

$$\Delta T \equiv T_N - T = DH^2, \qquad (1)$$

where $D = (1.65 \pm 0.15)\times 10^{-10}\,°\mathrm{K/G}^2$. Heller's low-field NMR data[6] give $D = (1.95 \pm 0.3)\times 10^{-10}\,°\mathrm{K/G}^2$. Calculations based on the molecular-field approximation indicate that at temperatures just below $T_N$, $\Delta T$ is in proportional to $H^2$. The proportionality constant $D$ is, however, 25 to 60% lower than the experimental value, depending on the procedure used in the calculation.[7]

A more satisfactory calculation of $D$ uses a thermodynamic relation derived by Skalyo et al.[8] This relation starts from Fisher's formula[9] which connects the magnetic contribution to the specific heat $C$, at $H = 0$, and the temperature dependence of the susceptibility $\chi_\parallel$ for $\vec{H}$ along the preferred axis, viz.,

$$C = A d(\chi_\parallel T)/dT, \qquad (2)$$

where $A$ is a slowly varying function of $T$ near $T_N$. Skalyo et al. have shown that near $T_N$ the P-AF boundary satisfies

$$d^2T/dH^2 = -A^{-1}. \qquad (3)$$

Using the specific heat data of Teaney[10] and the susceptibility data of Foner[11] we estimate $A = (3.2 \pm 0.3)\times 10^9\ \mathrm{G}^2/°\mathrm{K}$. Fisher's analysis[9] of older experimental data gives $A \cong 2.6\times 10^9\ \mathrm{G}^2/°\mathrm{K}$. Assuming that $\Delta T$ obeys Eq. (1) near $T_N$ and using our estimate for $A$ we obtain $D = (1.56 \pm 0.16)\times 10^{-10}\,°\mathrm{K/G}^2$, in good agreement with the experimental value.

It is interesting to compare the present results with those on other antiferromagnets with the cation $Mn^{++}$. For this purpose we define a reduced temperature $t = T/T_N$ and a reduced field

$$h = g\mu_B[S(S+1)]^{1/2}H/kT_N, \qquad (4)$$

where $\mu_B$ is the Bohr magneton, $g$ is the $g$ factor (2.0 for $Mn^{++}$), and $S$ is the spin quantum number (5/2 for $Mn^{++}$). Equation (1) can then be rewritten as

$$t = 1 - ah^2, \qquad (5)$$

where

$$a = k^2 T_N D/g^2\mu_B^2 S(S+1). \qquad (6)$$

Using relations given by Fisher[9] one can show that $a = r/12$, where $r$ is a factor of order unity.[12] Our results for $MnF_2$ give $a = 0.070 \pm 0.006$. The data of Schelleng and Friedberg[3] for $MnBr_2 \cdot 4H_2O$ give $a = 0.076$. Using the resonance results of Gijsman et al.[13] for $MnCl_2 \cdot 4H_2O$ one obtains $a \cong 0.063$, whereas their magnetization data give $a \cong 0.073$.

Turning to the AF-SF boundary, we have compared our results with the formula[11]

$$H_{sf} = [2H_E H_A/(1-\alpha)]^{1/2} \qquad (7)$$

for the spin-flop field $H_{sf}$. Here, $H_E$ is the exchange field, $H_A$ is the anisotropy field, and $\alpha = \chi_\parallel/\chi_\perp$ is the ratio of the susceptibilities for $\vec{H}$ parallel and perpendicular to the $c$ axis. Values for $2H_E H_A$ were taken from the resonance data of Johnson and Nethercot.[14] The coefficient $\alpha$ was evaluated using three sets of data.[11,15,16] At $T \lesssim 50$°K Eq. (7) gives values which are in agreement with experiment. The situation at $T \gtrsim 50$°K is less clear cut because of the sensitivity of the calculated value of $H_{sf}$ to small variations in $\alpha$ when $\alpha$ becomes comparable with unity. Using the susceptibility data of Ref. 11 one obtains from Eq. (7) a value for $H_{sf}$ which is about 13% lower than the experimental value at 60°K. Better agreement is obtained if $\alpha$ is deduced from the data of either Ref. 15 or Ref. 16. Theoretically one expects that Eq. (7) underestimates $H_{sf}$ near $T_3$, because in deriving Eq. (7) one neglects the increase in $\chi_\parallel$ with $H$. Calculations based on the molecular-field approximation show that the field dependence of $\chi_\parallel$ increases $H_{sf}$ by about 5% at

60°K, and by about 13% at 64°K. Unfortunately, in order to verify these deviations from Eq. (7) one needs more accurate data on $\alpha$ and $2H_EH_A$ than are available at present.

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# SEARCH FOR A POSSIBLE $I=0$, $Y=0$ BARYON IN $\bar{K}^-d$ INTERACTIONS AT LOW ENERGY*

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An attempt has been made to detect possible existence of a new neutral hyperon by studying both the missing-mass system and the $\Lambda\gamma$ system. Our result does not warrant the necessity of introducing any new hyperon near the mass of $\Lambda$, although it is not sensitive enough to dismiss the possibility entirely.

According to the SU(3) scheme, the existence of a singlet member belonging to the baryon nonet that contains the nucleons is possible, although its mass cannot be predicted. Even without the theoretical motivation it would still be interesting to search for any possible new $I=0$ hyperon near the $\Lambda$-$\Sigma$ mass region, hereafter referred to as $X^0$. An investigation in this region has not previously been reported. We have carried out a search for the possible existence of $X^0$ that might be produced in $K^-d$ interactions at low energy. In particular, we looked for it in the region just slightly above the mass of $\Lambda$. Our investigation includes the study of the missing-mass system as well as the search for the possible decay products $\Lambda+\gamma$ and/or $\Lambda+2\gamma$.

Approximately 80 000 pictures were taken to study $K^-d$ interactions by exposing the 30-in. Brookhaven National Laboratory deuterium-filled bubble chamber to a low-energy separated $K^-$