ANTIFERROMAGNETIC TO PARAMAGNETIC TRANSITION IN Fe\textsuperscript{2+}: MnF\textsubscript{2} IN EXTERNAL MAGNETIC FIELDS

C.R. Abeledo, Dept. of Chemistry, Brandeis University, Waltham, Massachusetts 02154

R.B. Frankel and M.A. Weber,\footnote{On leave from the Universidad de Chile, Casilla 5487, Santiago, Chile. Supported by the Organization of American States Fellowship.} Francis Bitter National Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

A. Misetich, Gerencia de Investigaciones, Comision National de Energia Atomica, Buenos Aires, Argentina

ABSTRACT

Mössbauer effect measurements across the antiferromagnetic to paramagnetic phase boundary in Fe\textsuperscript{2+}: MnF\textsubscript{2} in external magnetic fields are reported. From the data, $J(Mn-Fe) = -1.7$ cm$^{-1}$. The phase boundary is found to vary as $H^6$, as in pure MnF\textsubscript{2}.

MnF\textsubscript{2} crystallizes in a rutile structure with a tetragonal lattice. Below the Néel temperature $T_N = 67.4$ K, the magnetic properties of MnF\textsubscript{2} are well understood in terms of an ideal, two sublattice antiferromagnet with the spins aligned along the tetragonal c-axis. The phase diagram of MnF\textsubscript{2} in the H-T plane has been studied by Shapira and Foner\footnote{Supported by the Organization of American States Fellowship.} and is shown in Fig. 1. At low temperature, an external magnetic field applied along the c-axis causes a first-order realignment of the sublattice magnetization from along the c-axis to the basal plane when the magnitude of the external field reaches the critical value $H_{sf}$. At higher temperature, specifically above 65 K, the external field causes a second order transition to the paramagnetic phase, i.e., effectively lowers the Néel temperature.

Fe\textsuperscript{2+} may be isomorphously incorporated into the MnF\textsubscript{2} lattice. The effect of the addition of iron is to increase the Néel point\footnote{Supported by the Organization of American States Fellowship.} and to increase the value of $H_{sf}$. In a previous work, we observed the spin flop in Fe\textsuperscript{2+}: MnF\textsubscript{2} using Mössbauer spectroscopy in $^{57}$Fe\textsuperscript{4}. In this paper we report measurements of the antiferromagnetic to paramagnetic phase transition in external magnetic fields for single crystals of MnF\textsubscript{2} doped with 1% $^{57}$Fe.

A large single crystal of ~ 1% $^{57}$Fe\textsuperscript{2+} doped MnF\textsubscript{2} was grown from the melt by Optovac, Inc. The single crystal was oriented and a 6 mil slice was cut perpendicular to the c-axis and mounted between beryllium disks. In addition, some of the crystal was crushed and the powder was cast in lucite. Measurements were made in a conventional constant acceleration spectrometer operating in the normalized mode. The sample was placed in a cryostat which was inserted in a liquid nitrogen dewar which was in turn inserted into a superconducting solenoid operating in the persistent mode up to 85 kOe. The temperature was...
Fig. 1.
Phase diagram of pure MnF$_2$ from Ref. 1. Inclusion of Fe$^{2+}$ raises $H_{SF}$ and increase $T_N$. The measurements reported here are across the AF-P phase boundary.

Fig. 2.
Mössbauer spectra of single crystal Fe$^{2+}$: MnF$_2$ with $H_0 = 80$ kOe along the c-axis above and below $T_N$. 
controlled by pumping the liquid nitrogen bath below the desired temperature and then heating the sample electrically. The temperature was measured using a wire wound Pt resistor and the values were corrected for the effect of the external field using the results of Neuringer et al.\textsuperscript{5}

Because the field at the nucleus $\mathbf{h} = \mathbf{h}_{hf} + \mathbf{h}_0$, the spectra of the spin up and spin down sublattices in the antiferromagnetic phase are observed independently and the complete spectrum consists typically of eight lines (Fig. 2). In the paramagnetic phase all the spins are equivalent and the spectrum consists of just four lines (the $\Delta m=0$ lines are absent because the $\gamma$-ray propagation direction is parallel to $\mathbf{h}_0$). Thus we observe the temperature dependence of the spin up and spin down sublattices independently and the transition to the paramagnetic phase in clearly delineated.

![Graph](image)

Fig. 3. $|\mathbf{h}_{hf}|$ and $\mathbf{h}_{hf}/\mathbf{H}_{sat}$ plotted as a function of $T$ for $H_0=80$ kOe.
The data were analyzed using a computer program of Singh and Hoy. From the quadrupole splitting above $T_N$ and powder spectra at several points below $T_N$ we found that the quadrupole coupling parameter $Q = 2.95$ mm/sec. and the asymmetry parameter $\eta = 0.4$ did not change appreciably in the transition region.

In Fig. 3, we plot the hyperfine field $H_{hf}$ in the Fe$^{2+}$ in the spin down and spin up sublattices as a function of temperature for an applied field $H_0 = 80$ kOe. The data have been analyzed using a molecular field approximation model where $J(Mn)$ and $J(Fe)$, the Mn-Mn and Fe-Fe exchange constants respectively were chosen to give the best fit to the Néel point of pure MnF$_2$ and pure FeF$_2$. The Fe$^{2+}$ single ion anisotropy constant $D$ was taken from the measurements of Lowe et al.; $D = 8.6$ cm$^{-1}$. Using these values the best fit to the Néel point is obtained with $J = -1.5$ cm$^{-1}$, however the best fit to the magnetization above and below $T_N$ is obtained with $J = -1.7$ cm$^{-1}$.

![Fig. 4. $H^2$ plotted as a function of T for MnF$_2$ and Fe$^{2+}$:MnF$_2$](image-url)
Shapira and Foner\(^1\) and Heller\(^6\) showed that the phase boundary in pure MnF\(_2\) is well represented by an equation of the form \(T - T_N = AH^2\), where \(A\) is a constant. In Fig. 4 we plot \(T_N\) determined from our data as a function of \(H^2\), including one value obtained by holding \(T\) constant and changing \(H_0\). The boundary is seen to vary as \(H^2\) as in MnF\(_2\) and with the same slope. This result is predicted by the M.F.A.


