

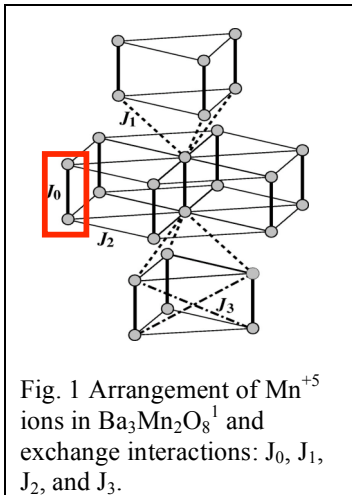
Exploring the Exotic Phases of an S=1 spin system: Ba₃Mn₂O₈

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Abstract

Ba₃Mn₂O₈ is an S=1 coupled spin dimer with a gapped ground state¹. Both magnetization as a function of magnetic field at a temperature of 0.65K and specific heat data suggest exotic phase transitions in the system. This paper presents the theoretical framework and some experimental considerations for an experiment using nuclear magnetic resonance (NMR) to study the structure of these exotic phase transitions in the Ba₃Mn₂O₈ system.

Introduction



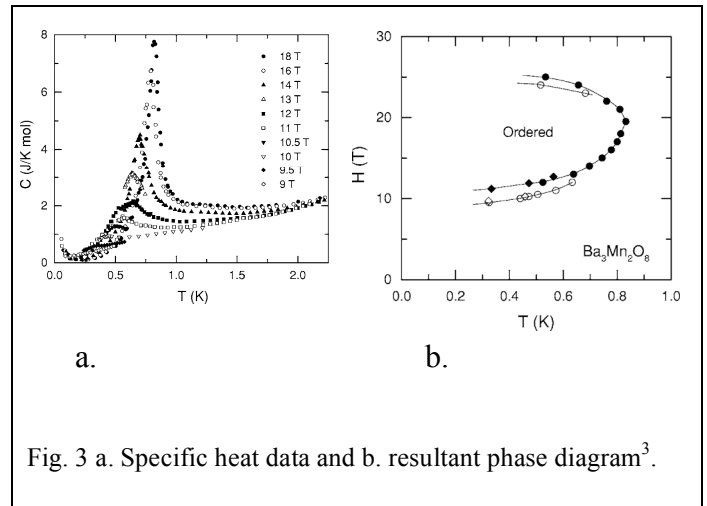
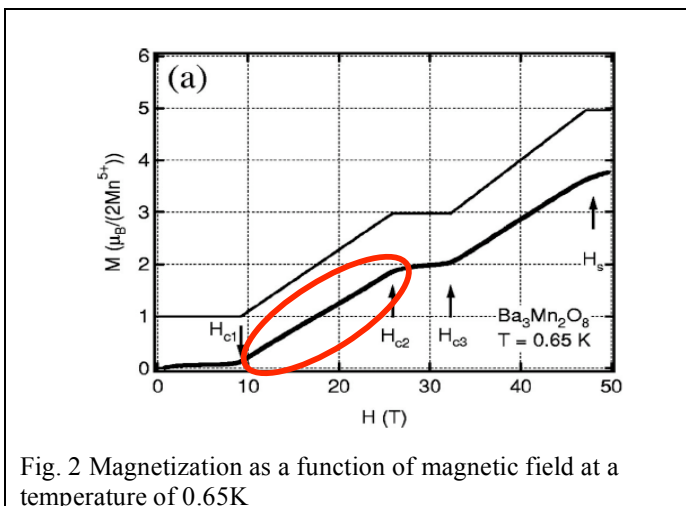
Finding macroscopic examples of quantum phenomena is currently of great interest in modern physics. One material that demonstrates macroscopic quantum behavior is Ba₃Mn₂O₈, an S=1 coupled spin dimer with a gapped ground state. The spin for this system is carried by the Mn⁵⁺ ion.

Fig. 1 shows the arrangement of Mn⁵⁺ ions in Ba₃Mn₂O₈. The red region highlights a pair of highly interacting spins that make up the strongly coupled pair of spins called a dimer. This is indicated by the strong intradimer exchange interaction J₀ compared to the interdimer exchange interactions: J₁, J₂, J₃. A look at magnetization as a function of magnetic field at a temperature of 0.65K shows plateaus and sloped regions (Fig. 2)¹. This is due to the lowering of energy for excited states as field is increased. For fields less than H_{c1}, the singlet state, |00>, dominates. At H_{c1} the energy gap between the singlet and triplet states closes, and so for H_{c1}<H<H_{c2} there is a mix of

states in the |0,0> state and the |1, S^z> where S^z=1, -1 states. As the field approaches H_{c2} the number of dimers in the |1, 1> state saturates. The |1,1> state dominates for H_{c2}<H<H_{c3}. In the region H_{c3}<H<H_s it again becomes energetically favorable for higher excited states to be present. In this region a mix of the |1,1> state and |2, S^z> with S^z=2,0 states are present. As field is increase to H_s the |2,2> state become the most prevalent. After a field of H_s, saturation is again attained with nearly all dimers in the |2,2> state.

Interestingly, regions where there is a mix of states suggest the phenomena of a magnetic version of Bose-Einstein condensation (BEC)². For instance, in the region H_{c1}<H<H_{c2}, as the field is increased, more and more dimers enter the |1,1> state. By changing the field one can then tune the number of |1,1> states present. The condensation of |1,1> states can be understood as caused as a Bose-Einstein condensation brought on by magnetic field. The same considerations can be made for the region H_{c3}<H<H_s except that the state that can be tuned with the field in that region is the |2,2> state.

BEC implies that there is some sort of symmetry breaking occurring. The fact that symmetry is breaking indicates a change of phase in the system where the symmetry is breaking. Specific heat data confirms phase transitions as suggested by peaks in specific heat versus temperature (Fig. 3a)³.



This specific heat data was taken to produce the phase diagram shown in Fig. 3b³. The data show that there are two distinct phase transition occurring at temperatures below 1K and between magnetic fields of 9T and 26T.

The purpose of this experiment is to use NMR to gain information about the structure of these exotic phases. Although the Mn⁺⁵ ion carries the spin, the Ba nucleus will be what will be studied using NMR. This is possible due to the close proximity of the to nuclei. In other words, information about the Ba nucleus yields information about the Mn nucleus, which in turn yields information about the electron structure of the exotic phases.

NMR Theory⁴
Zeeman effect

According to the Zeeman effect, an applied magnetic field introduces an extra term in the spin Hamiltonian of:

$$H_{ZE} = - \boldsymbol{\mu} \cdot \mathbf{H}$$

where $\boldsymbol{\mu}$ is the magnetic moment and \mathbf{H} is the magnetic field. Hence a magnetic field adds H_{ZE} to the spin Hamiltonian, breaking the degeneracy between spin states of different z component of spin angular momentum (Fig 4).

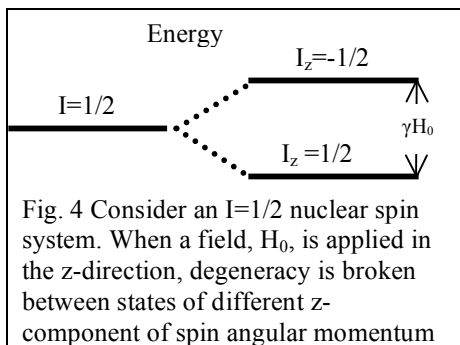


Fig. 4 Consider an I=1/2 nuclear spin system. When a field, H₀, is applied in the z-direction, degeneracy is broken between states of different z-component of spin angular momentum

To understand the importance of this fact to NMR, first consider the nuclear magnetic moment. The magnetic moment is proportional to the nuclear spin quantum number, \mathbf{I} :

$$\boldsymbol{\mu} = \gamma \mathbf{I}$$

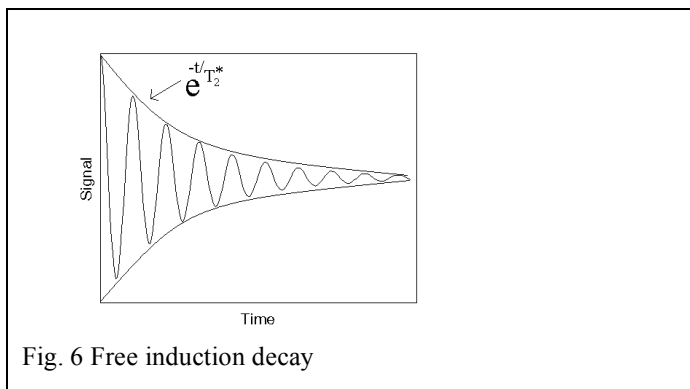


Fig. 6 Free induction decay

where γ is the gyromagnetic ratio. The z- component of the nuclear spin can take on values of $m\hbar$ where m can range from $-I$ to $+I$, leading to $2I+1$ possibilities. The magnetic moment in the z-direction is $m\gamma\hbar$. The energy shift caused by H_{ZE} is $E = \gamma H_0 \hbar m$ and comparison with Planck's equation, $E = \omega \hbar$, yields that sending in photons with angular frequency $\omega = \gamma H$ (called the Larmor angular frequency) will excite the nucleus to a spin state higher in energy. For Ba, γ is equal to $2.993 \cdot 10^7$ radians / (s·T).

In practice, to supply the energy necessary to excite transitions, an oscillating voltage pulse with frequency centered at the Larmor frequency is transmitted through a coil that surrounds the sample. The oscillating voltage creates an oscillating current, which induces a changing magnetic field and thus provides an oscillating field with the Larmor frequency.

Magnetization

The static field tries to line up moments with the field. This produces a net magnetization. Classically, a moment in a static magnetic field produces a torque, $\boldsymbol{\tau} = \boldsymbol{\mu} \times \mathbf{H}_0$ and has the effect of causing the magnetization to precess about the magnetic moment. The precession frequency is given, again, by the Larmor relation $\omega = \gamma H_0$.

Applying a magnetic field that oscillates at the Larmor frequency in the plane perpendicular to the static field H_1 can cause the nuclear moments to undergo transitions to higher energy states. These transitions will cause the entire macroscopic magnetization to turn towards or away from the static field.

Reference Frames

It is convenient to consider a frame of reference rotating at the Larmor frequency. The rotating reference frame with axis x' , y' , and z' is related to the laboratory frame with axis x , y , z by:

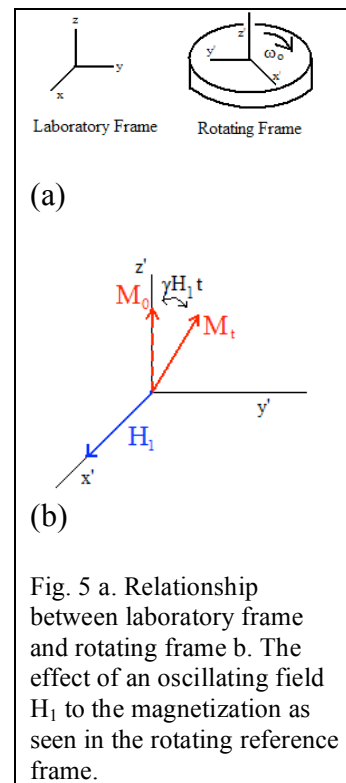


Fig. 5 a. Relationship between laboratory frame and rotating frame b. The effect of an oscillating field H_1 to the magnetization as seen in the rotating reference frame.

equal to z' , and x' and y' rotate with respect to x and y . In this frame the magnetization is static (Fig. 5). Then by applying a magnetic field that oscillates with the Larmor frequency, $H_1 \cos \omega t$ in say the x' direction, the magnetization, \mathbf{M} , will rotate in the rotating frame about this H_1 field at angular speed $\omega = \gamma H_0$ onto say the y' direction. Since H_1 is perpendicular to the laboratory field H_0 , the magnetization will change its orientation with respect to the static field as shown in Fig. 5b. Thus, the appropriate combination of

intensity and duration of the rotating field can rotate the magnetization by any desired amount. After the rf field H_1 is turned off, the magnetization will precess freely but decay over time. In a coil with its axis perpendicular to H_0 , this decaying magnetization will induce a current that oscillates with the Larmor frequency allowing a signal to be picked up and converted to a voltage. This signal induced in the coil is called the free induction decay (FID) signal and is depicted in Fig. 6.

Relaxation Times

Certain characteristic times after a rf pulse is applied are important in NMR. First consider T_2^* . It describes the decay rate of the magnetization in the $x'-y'$ plane given by e^{-t/T_2^*} . This decay is partly due to the fact that there may be local differences in the field that each nucleus feels and hence different section of a material may have moments that precess at slightly different Larmor frequencies. The nuclear moments may then dephase, causing the net magnetization to decrease. When these differences in field are removed one is left with the intrinsic decay rate given by T_2 , the spin-spin relaxation time. T_2 is related to T_2^* by

$$\frac{1}{T_2^*} = \sqrt{\left(\frac{1}{T_2}\right)^2 + (\gamma \Delta H_0)^2}$$

Another characteristic time is the spin-lattice relaxation time, T_1 . This time gives a measure of the time it takes for the magnetization to return to equilibrium through the equation:

$$M_z = M_0 [1 - e^{-t/T_1}]$$

Fig. 7 shows the magnetization after applying a rf pulse in the x' -direction that knocks the magnetization to the y' -direction.

Hyperfine Coupling

NMR allows one to infer information about phase through the hyperfine coupling which adds an extra term to the nuclear spin Hamiltonian:

$$H_{HC} = a \mathbf{I} \cdot \mathbf{S}$$

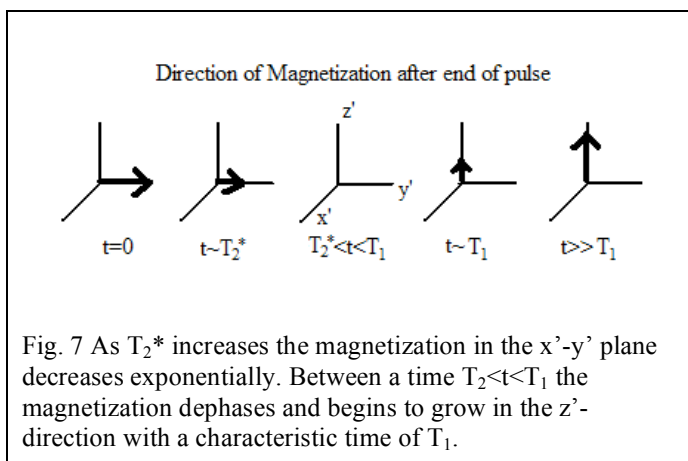
where \mathbf{I} is the nuclear spin and \mathbf{S} is the electronic spin. For example, a manifestation of hyperfine

$$K = \frac{\Delta H}{H_0} = \frac{a \chi_s}{g N \mu_B \gamma \hbar}$$

coupling is the Knight shift:

which is dependant on the coupling term, a , and for a given material changes with spin susceptibility. This coupling actually allows for understanding the phase of a material. Next, consider an antiferromagnetic material with a critical temperature T_N . Above T_N the material is in its paramagnetic state with spins randomly aligned and shifts may be seen given by the knight shift. Below T_N the spins are aligned so that each spin is surrounded by nearest neighbors of opposite spin. This may cause, say, the spin ups to give H_{HC0} whereas the spin downs may give the opposite value, and thus causing different shifts in energy. This causes the energy absorption line to split in half giving two absorption lines.

Thus, by looking at NMR data in the region where the new phases are believed to exist will give information about the spin structure and hence the nature of the phase.



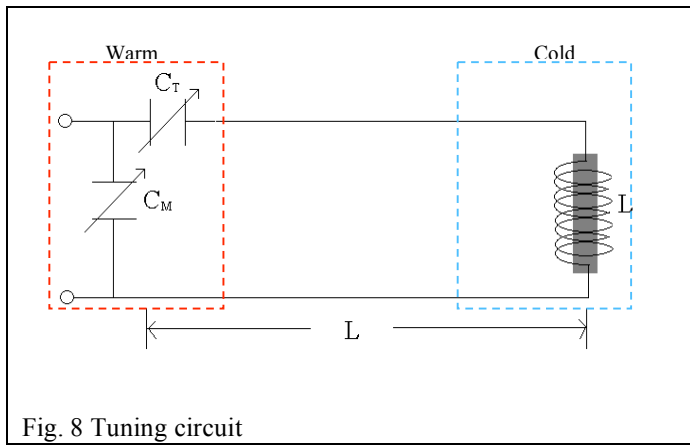


Fig. 8 Tuning circuit

Experimental setup

Tuning Circuit

In order to provide the rf pulse with center frequency being the Larmor frequency, a modified LC circuit is used which has a resonance condition of:

$$\omega = \frac{1}{\sqrt{C_T L}}$$

However there is also an additional capacitor that is put in parallel with the capacitor, C_T , and the inductor, L . This additional capacitor, C_M , is a matching capacitor that help the circuit match the impedance of the cable, 50Ω . Another modification to the traditional LC circuit analysis is that the finite distance between the capacitors and the inductors. When this distance becomes comparable to the wavelength of the signal being transmitted, other resonance conditions are created. One of these resonance conditions must equal the Larmor frequency in order for energy to be supplied to the sample. Fig. 9 shows various resonance conditions for a finite length of cable.

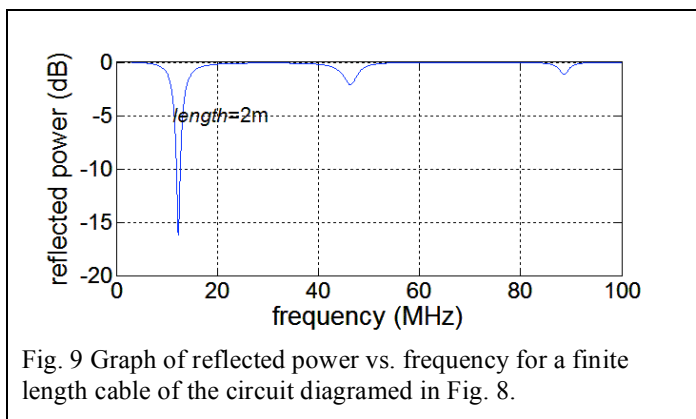


Fig. 9 Graph of reflected power vs. frequency for a finite length cable of the circuit diagramed in Fig. 8.

Preliminary Data

Fig. 10 shows a graph of absorption vs the difference between the frequency sent in which is determined by the Larmor frequency and the frequency of the FID signal. The data was taken at a temperature of 1.5K and at a magnetic field of 8.5T. Without the hyperfine effect the Larmor frequency would equal the FID frequency and hence the peak

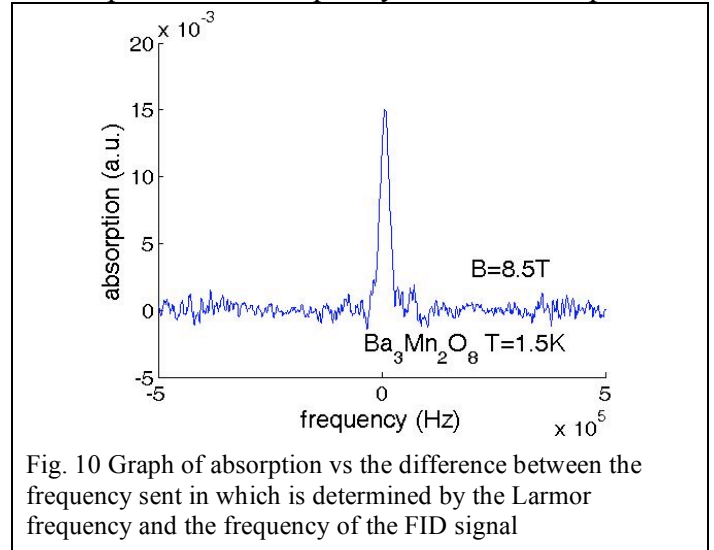


Fig. 10 Graph of absorption vs the difference between the frequency sent in which is determined by the Larmor frequency and the frequency of the FID signal

would be centered on 0Hz. As the graph shows the peak in absorption does not occur 0Hz but rather off zero and hence demonstrates the effect of hyperfine coupling. By going down to the temperature and magnetic field region in which the exotic phases are present, NMR will give information about the electron structure.

Conclusion

NMR provides a means to study these exotic phase transitions. Once the dilution refrigerator, which allows one to reach temperature on the order of mK, is operation data can be taken.

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