

Spin state of Mn_3O_4 investigated by ^{55}Mn nuclear magnetic resonance

2012 March Meeting, Boston, MA

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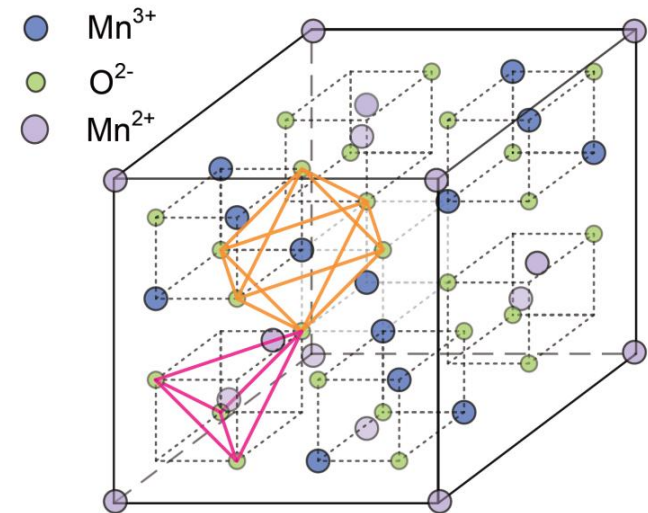
PRB 84 174423 (2011)

Outline

- Structural and magnetic properties of the spinel oxide Mn_3O_4
- Nuclear magnetic resonance experiments for the magnetic materials
- Spin state of Mn_3O_4 investigated by NMR

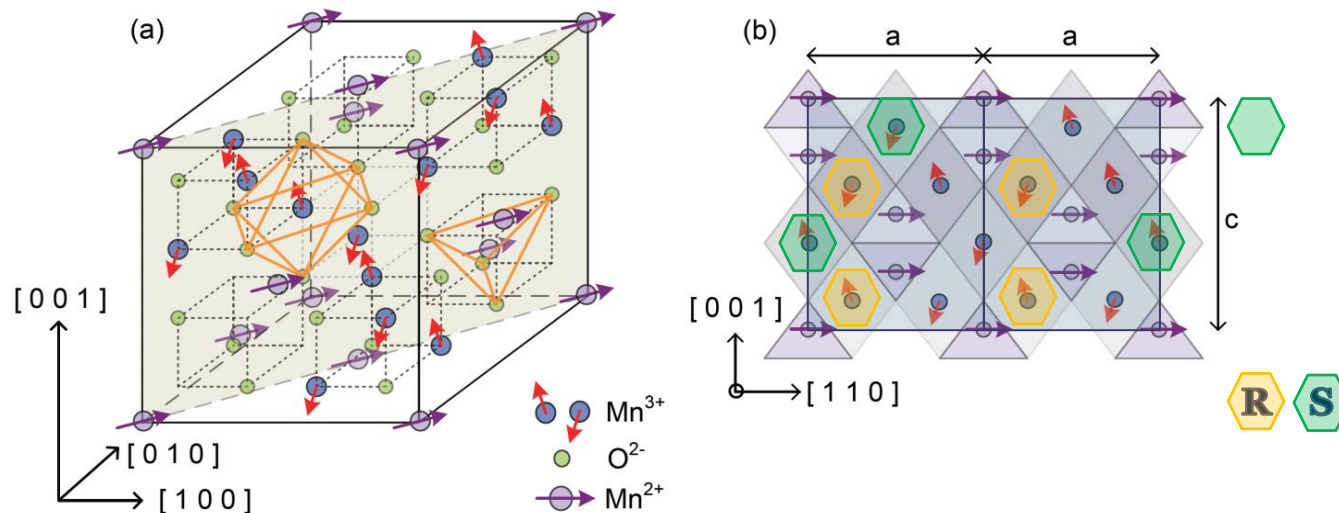
Properties of the spinel oxide Mn_3O_4

- AB_2O_4 spinel structure
- Mn^{2+} ($3d^5$) ion in the A(tetrahedral) site
- Mn^{3+} ($3d^4$) ion in the B(octahedral) site
- Phase transition



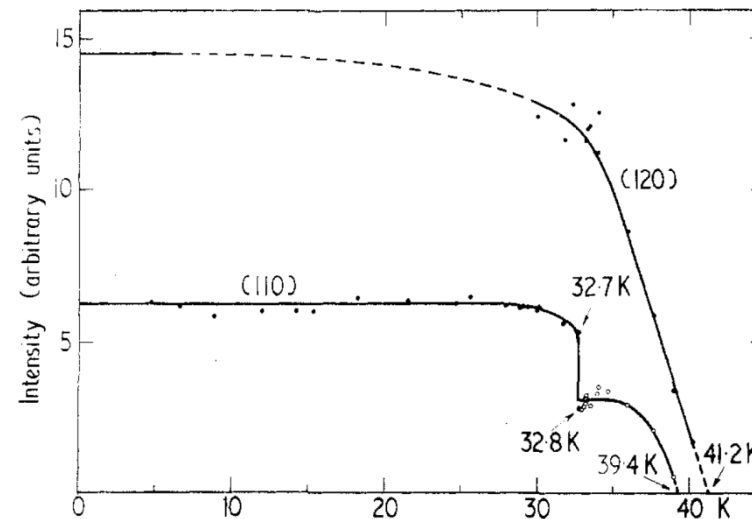
Structural	monoclinic	tetragonal		cubic
Magnetic phase	Cell doubled phase(ferri-)	Incommensurate conical phase	Yafet-Kittel(ferri-)	Paramagnetic phase
	33K	39K	43K	1443K

Cell doubled phase



- Commensurate ferrimagnetic ordering in which the net magnetic moment is along the [110] direction
- The magnetic unit cell doubles the chemical unit cell

Commensurate-Incommensurate phase transition



[J. Phys. C. (1974) G. B. Jensen and O. V. Nielsen]

- Different temperature dependence of neutron intensity for (120) reflection related to the R spin and (110) reflection related to the S spin
- Only S spin (cell doubling spin) is related to the Commensurate-Incommensurate phase transition

Different magnetic moment R and S

- According to the B. Boucher, R. Buhl and M. Perrin's neutron diffraction experiment results, the magnetic moment R and S have **the same magnitude** but **different canting angles**

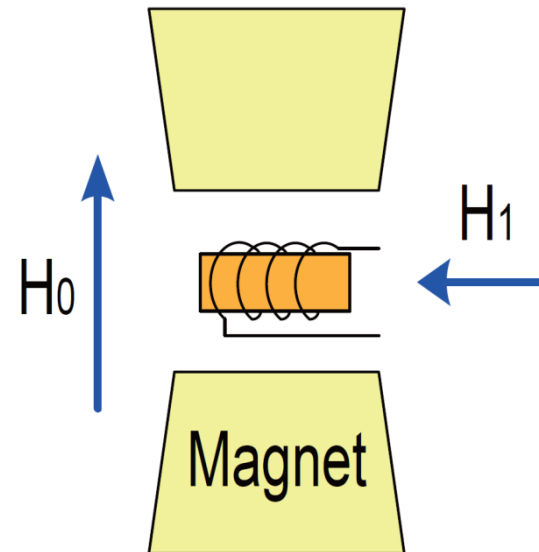
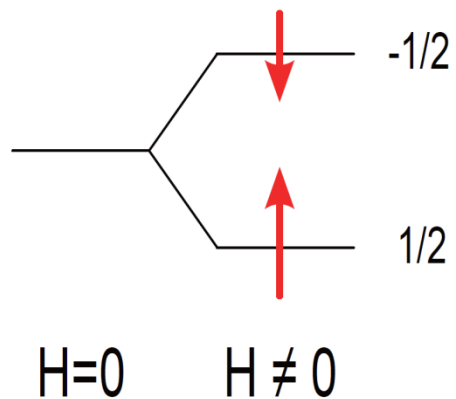
[B. Boucher, R. Buhl, and M. Perrin, J. Appl. Phys. **42**, 1615 (1971)]

- According to the G. B. Jensen and O. V. Nielsen's neutron diffraction experiment results, the magnetic moment R and S have **the same canting angle** but **different magnitudes**

[G. B. Jensen and O. V. Nielsen, J. Phys. C **7**, 409 (1974)]

NMR (nuclear magnetic resonance)

- Good technique to investigate magnetic properties as a local probe



$$\omega = \gamma H$$

Zero field NMR

- In general:

$$\omega = \gamma H \rightarrow \omega = \gamma(H_{hyp} + H_{ext})$$

- In absence of the external magnetic field :

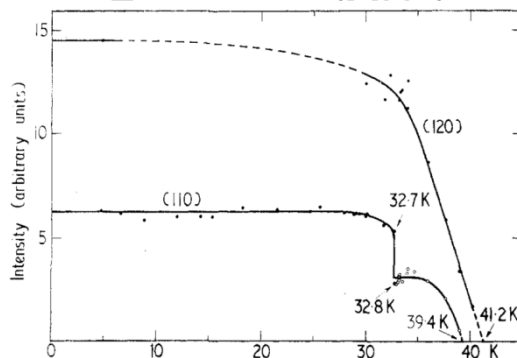
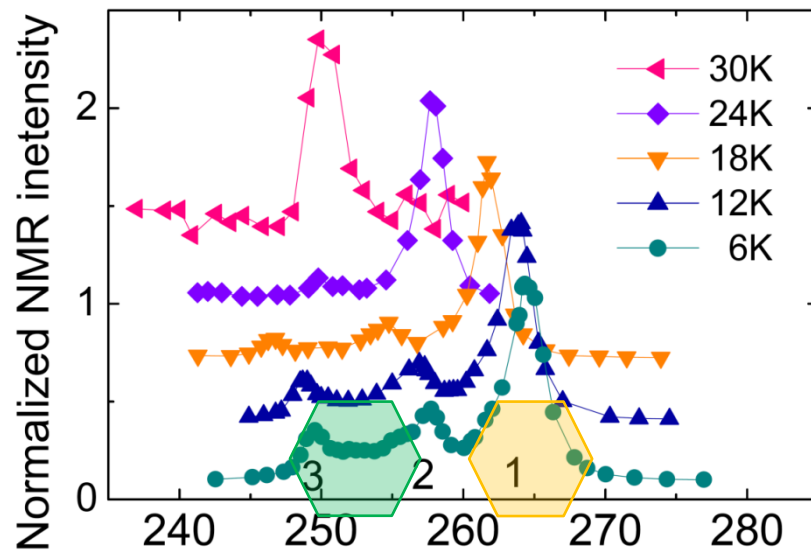
$$\omega = \gamma H_{hyp}$$

- In the magnetic material :

$$\omega = \gamma A \mu, \text{ (A = hyperfine field constant, } \mu = \text{ magnetic moment)}$$

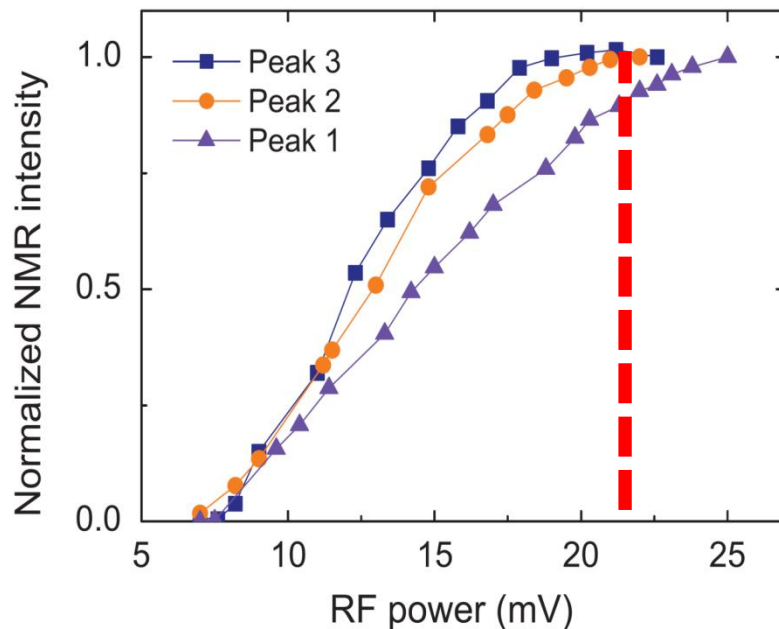
- Resonance frequency is proportional to the magnetic moment !!

Temperature dependence of the NMR spectrum



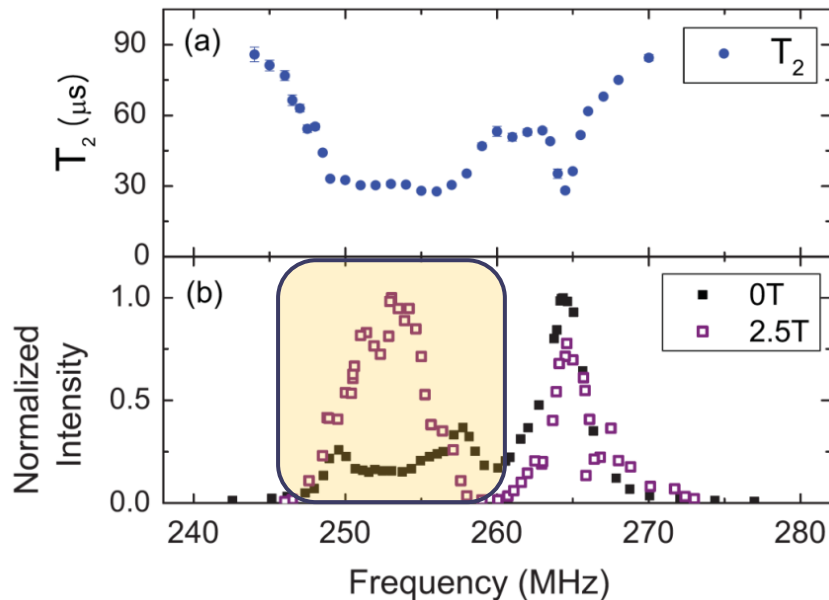
- Zero field NMR spectrum for Mn³⁺ ion in Mn₃O₄ consists of 3 peaks
- The temperature dependence of the each peak is different
- The peak1 comes from R spin and peaks 2 and 3 come from S spin which is related with commensurate-incommensurate phase transition

RF power dependence of peak1,2 and 3



- Different rf power dependence of the normalized nmr intensity at each peak position
- rf enhancement created by oscillating electron spin depends on the anisotropy field
- Magnetic moment related to the peak 1 is in the different environment from that of peak 2 and 3

Suhl-Nakamura interaction in Mn_3O_4



- Frequency dependence of the Spin – spin (T_2) relaxation time
- Central parts of the peaks 2 and 3 in the spectrum measured in 0T magnetic field is suppressed
- The fast relaxation at the peak position due to the **Suhl-Nakamura interaction**
- Spin – spin relaxation due to Suhl-Nakamura interaction gets slower with external magnetic field
- Suhl-Nakamura interaction broadens the peak related to the magnetic moment S

Conclusions

- The temperature dependence of the NMR spectrum and the rf enhancement factor show that Mn^{3+} ions have **two different states of magnetic moment**, one of which is strongly related to the **commensurate-incommensurate phase transition**
- The Suhl-Nakamura interaction is the one of the main relaxation mechanism