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*The Organizing Committee of the XXVI
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I. Bakaimi

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titled 'Frustration and Induced
Magnetodielectric Coupling in NaMnO₂
Polymorphs'*

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Frustration and Induced Magnetodielectric Coupling in NaMnO_2 Polymorphs.

Ioanna Bakaimi^{1,2*}, Othon Adamopoulos^{1,3}, Bohdan Kundys⁴, Mark A. Green^{5,6},
Chris Stock⁵, and Alexandros Lappas¹

¹*Institute of Electronic Structure and Laser, Foundation for Research and Technology - Hellas,
Vassilika Vouton, 71110 Heraklion, Greece*

²*Department of Physics, University of Crete 71003, Heraklion, Greece*

³*Department of Chemistry, University of Crete 71003, Heraklion, Greece*

⁴*Service de Physiques de l'Etat Condense, CEA Saclay, 91191 Gif-Sur-Yvette Cedex, France*

⁵*NIST Center for Neutron Research, 100 Bureau Drive, Gaithersburg, Maryland 20899-8562, USA*

⁶*Department of Materials Science and Engineering, University of Maryland, College Park, Maryland
20742-2115, USA*

*bakaimi@iesl.forth.gr

The hallmark of frustrated magnets in physics of materials is attributed to two reasons: on the fundamental side it is possible to understand how the lattice topology imposes the formation of a simple, ordered spin configuration [1], on the applied side the instabilities that are observed in frustrated magnets are promising for the development of novel materials with numerous applications.

The polymorphs α - NaMnO_2 (Fig.1a) and β - NaMnO_2 (Fig.1b) belong to the most simple case of frustrated antiferromagnets due to their triangular magnetic lattices. In a triangular topology each spin cannot satisfy all pairwise interactions and therefore magnetic disorder remains at temperatures well below the Curie-Weiss temperature. This leads to a degenerate ground state. However, long range magnetic order is possible to occur by modifying the topology of the lattice and lifting of the degeneracy.

The goal of this work is twofold. First to add knowledge to the mechanisms that raise the geometrical frustration in a series of compounds with triangular topology and lead to LRO (long range order) and second to reveal the mechanism that underlies in the induced magnetodielectric coupling in such antiferromagnets. An example of the former is found in α - NaMnO_2 while the β - NaMnO_2 is explored for the latter behavior. Understanding the impact of the topology in their magnetic properties is of prime importance.

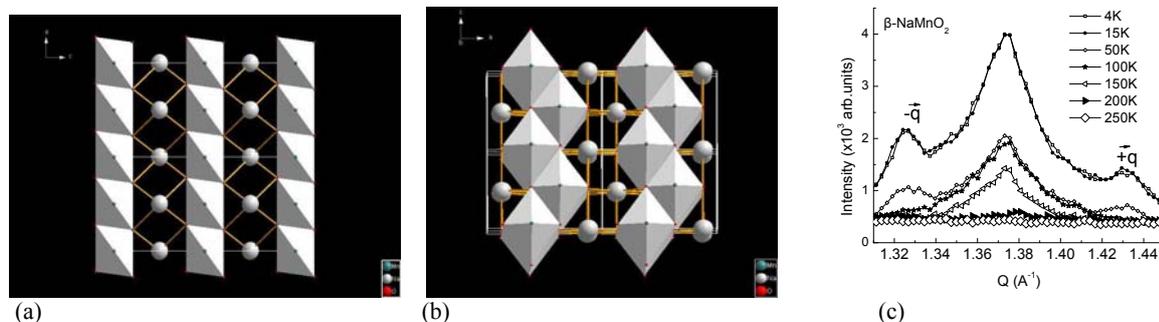


Figure 1. Crystal Structures (projections on the a-c plane) (a) α - NaMnO_2 , (b) β - NaMnO_2 and (c) portion of neutron powder diffraction data that shows the main magnetic Bragg peak and its satellites for β - NaMnO_2 .

The manganese oxides were prepared by high temperature solid state reactions. The purity of the samples and the crystal structure was confirmed by X-Ray powder diffraction. Static magnetic ordering was investigated by high resolution NPD (Neutron Powder Diffraction, at the BT1 diffractometer, $\lambda=2.0787\text{\AA}$ at NIST. Temperature evolution of the dc magnetic susceptibility was carried out at IESL-FORTH with a commercial SQUID magnetometer $5\text{ K} < T < 300\text{ K}$ ($H=0.2\text{ T}$).

Previous studies have proved that α - NaMnO_2 is a triangular antiferromagnet with anisotropic interactions ($J_1, J_2 < 0$) where the relief of frustration is connected with a structural transition from monoclinic (C2/m) to triclinic (P-1) symmetry [2]. The question that arises is whether an analogous symmetry breaking mechanism dictates the magnetic ground state of the β - NaMnO_2 polymorph?

Both of these oxides exhibit a layered structure where the Na^+ cations are separating the MnO_2 sheets. α - NaMnO_2 in Fig.1a: monoclinic, C2/m, $a=5.67\text{\AA}$, $b=2.86\text{\AA}$, $c=5.80\text{\AA}$, $\beta=113.14^\circ$ and the β - NaMnO_2 structure in Fig.1b: orthorhombic, Pmmn, $a=4.78\text{\AA}$, $b=2.86\text{\AA}$, $c=6.33\text{\AA}$. However in α - NaMnO_2 the MnO_2 sheets are built

up of flat edge sharing octahedra, whereas in β -NaMnO₂ the MnO₂ layers are built up of double stacked corrugated edge sharing MnO₆ octahedra [3]. Their local environment is dominated by the spontaneous distortion of the the Jahn-Teller active Mn³⁺ cation in octahedral geometry.

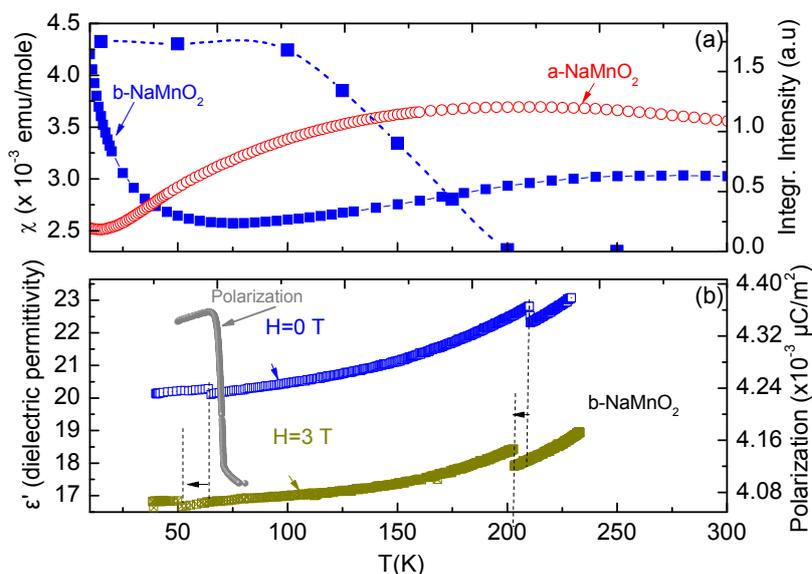


Figure 2. (a) Left y-axis. The dc magnetic susceptibility of α -NaMnO₂ and β -NaMnO₂ frustrated magnets (H=200 Oe, ZFC) Right y-axis: The integrated intensity of the strongest magnetic Bragg peak that develops below 200 K and indicates the development of a commensurate magnetic structure. (The line is a guide to the eye.)

(b) Left y-axis: Anomalies in dielectric permittivity of β -NaMnO₂ at 210 K and 65 K under H=0 T and H=3 T. Right y-axis: Polarization of β -NaMnO₂ around 65 K.

In view of the previous studies on the α -phase we present new results for the β -phase. Fig.1c, Fig. 2a and Fig.2b reveal the same similarities in the transitions witnessed by different techniques such as NPD, SQUID, and magnetodielectric measurements. The comparison of the data shows:

a) Similarity in the magnetic susceptibility of the two compounds: the broad hump at high-T which is a characteristic for developing 2D spin correlations. However the hump is shifted for the β -phase at much higher-T indicating that the strength of the magnetic interactions is enhanced.

b) Surprises that arise from the comparison of the transitions that appear in the NPD data (Fig.1c) and the dc susceptibility of β -NaMnO₂ (Fig.2a). Despite the fact that the dc susceptibility does not reveal clearly any phase transition the NPD data show that the first commensurate magnetic peak appears around 200 K and the satellite magnetic peaks start to emerge at the temperatures between 50 K-100 K. These latter peaks are in support of an incommensurate magnetic structure at low temperatures.

c) Anomalies in dielectric permittivity, ϵ' , versus temperature and zero magnetic field show two sharp changes: the first around T=200 K, and the second one at T=65 K. Importantly when one measures polarization, P, versus temperature an enhancement of P is found at 65 K, which indicates some degree of magnetodielectric coupling. Furthermore when a magnetic field of 3 T is applied there is an important shift to lower temperatures for both of the observed dielectric anomalies.

Further experiments are underway in order to examine the existence of possible ferroelectricity and the origin of the observed magnetodielectric coupling in the β -NaMnO₂ polymorph.

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