Incommensurate Magnetic Phases of the Multiferroic Compound $MnCr_2O_4$ Described with the Super-space Formalism

Miguel PARDO-SAINZ[1,2], Ayaka TOSHIMA[3], Gilles ANDRÉ[4], Juan BASBUS[5], Gabriel CUELLO[6], Takashi HONDA[7], Toshiya OTOMO[7], Katsuya INOUE[3], Yusuke KOUSAKA[2], Javier CAMPO[1,*]

[1]Instituto de Nanociencia y Materiales de Aragón (INMA), CSIC-University of Zaragoza, Zaragoza, Spain; [2] Osaka Prefecture University, Sakai, Japan; [3]Chirality Research Center and Institute for Advanced Materials Research, Hiroshima, Japan; [4]Laboratoire Léon Brillouin, Saclay, France; [5]Centro Atómico Bariloche, S. C. de Bariloche, Argentine; [6]Institut Laue-Langevin, Grenoble, France; [7]Institute of Materials Structure Science, Tsukuba, Japan

Introduction

❖ Nowadays, chromium-based normal spinel oxides ACr_2O_4 are one of the most studied materials in the condensed matter community due to the interplay between its magnetic, electric and structural properties [1,2].

❖ In particular, for $MnCr_2O_4$, the ground state magnetic structure is still controversial because the magnetic structures reported by different groups and investigated by independent techniques are inconsistent [1-3].

Super-space Group Formalism

❖ Incommensurate structure = basic structure + modulations:

$$\vec{M}_j(x_4) = \vec{M}_{j,0} + \sum_{n=1}^{\infty}\left[\vec{M}_{j,ns}\sin(2\pi n x_4) + \vec{M}_{j,nc}\cos(2\pi n x_4)\right]$$

❖ Symmetry operations: space group operations + phase shifts of modulations. Determined by the magnetic super-space group.

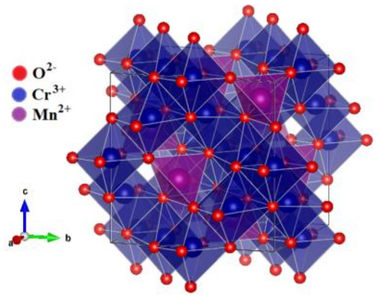
Methods

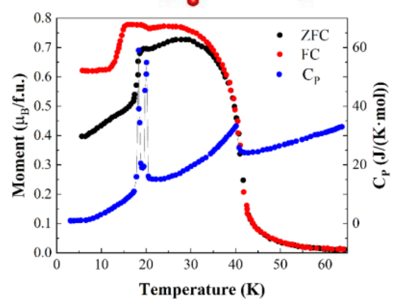
❖ The magnetic structure of this compound was reinvestigated by magnetization, specific heat and neutron diffraction at different temperatures.

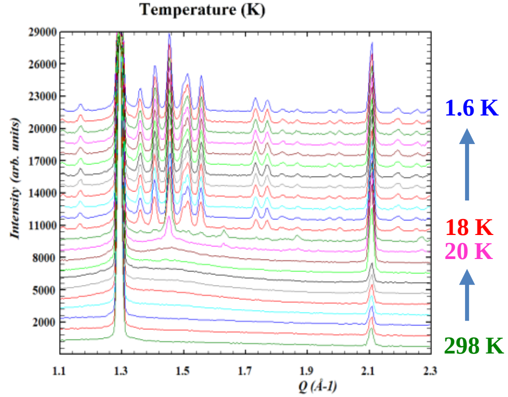
❖ The results suggested that a new magnetic phase, not previously reported, is developed under 18 K.

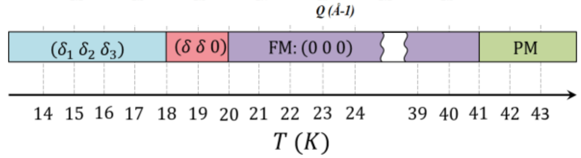
❖ The magnetic phases in this sample were:

- ➢ Ferrimagnetic order below T_C = 45 K
- ➢ Conical spin order with propagation vector $\vec{k}_{S1}$= (0.62(1), 0.62(1), 0) below T_{S1} = 20 K
- ➢ Conical spin order with propagation vector $\vec{k}_{S2}$ = (0.660(3), 0.600(1), 0.200(1)) below T_{S2} = 18 K.









Results

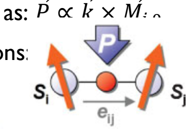
❖ Using the super-space group approach [4], the symmetry of the nuclear and magnetic structures is determined:

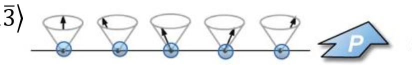
Phase	Group	Irr. Rep	$\vec{k}$	$\vec{M}_{j,0}$	$\vec{M}_{j,s}$	$\vec{M}_{j,c}$
PM	Fd-3m (#227)	-	-	-	-	-
FiM	Imm'a' (#74.559)	mGM_4^+	(0 0 0)	$\langle 1\bar{1}0\rangle$	-	-
δ δ 0	Im'a'2(0,0,g)0ss (#46.1.12.4.m245.1)	$mGM_4^+ \oplus mSM2$	(0.62 0.62 0)	$\langle 1\bar{1}0\rangle$	$\langle 110\rangle$	$\langle 001\rangle$
$\delta_1\delta_2\delta_3$	P1(a,b,g)0 (#1.1.1.1.m1.1)	$mGM_4^+ \oplus mGP1$	(0.66 0.6 0.2)	$\langle 100\rangle$	$\langle 010\rangle$	$\langle 001\rangle$

❖ Electric polarization $\vec{P} \propto \vec{r}_{ij} \times (\vec{S}_i \times \vec{S}_j)$, also can be expressed as: $\vec{P} \propto \vec{k} \times \vec{M}_{j,0}$

❖ Non-zero value of polarization for transverse conical modulations:

- ➢ δ δ 0 phase: $\vec{P} \parallel \langle 001\rangle$
- ➢ δ_1 δ_2 δ_3 phase: $\vec{P} \parallel \langle 01\bar{3}\rangle$





Conclusions

❖ New magnetic phase, not previously reported, identified under 18 K.

❖ Using SGF, symmetry of nuclear and magnetic structures is determined.

❖ Presence of transverse conical magnetic structures in lower-temperature phases implies existence of multiferroicity.

❖ Through simple theoretical calculations, we derive the macroscopic electric polarization vector for each magnetic phase.

References

[1] K. Dey et al., Journal of Magnetism and Magnetic Materials 435, 15 (2017).

[2] K. Tomiyasu et al., Phys. Rev. B 70, 214434 (2004).

[3] J. M. Hastings and L. M. Corliss, Phys. Rev. 126, 556 (1962).

[4] J. Rodríguez-Carvajal and J. Villain, Comptes Rendus Physique 20, 770 (2019).