

# Spontaneous Physical Property Tables for Single Crystals (1-x)Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - xPbTiO<sub>3</sub> (PMN-PT) in Multi-Ferroic Phase Transitions

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## Abstract

The crystals (1-x)Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - xPbTiO<sub>3</sub> (represented as (1-x)PMN-xPT) has a prototype phase Pm $\bar{3}$ m (underline parent point group is  $\bar{m}3m$ ) space group symmetry. In the range of temperatures 32 °C to 250 °C it has three phase transitions  $\bar{m}3mF4mm$ ,  $\bar{m}3mF3m$ , and  $\bar{m}3mFmm2$ . Based on the underline point group, the spontaneous property tables are constructed to identify the existence and changes in different physical properties that are exhibited by the crystal in each of the phase transitions.

**KEY WORDS:** Phase transitions, Spontaneous physical property, Ferroic properties, Poin groups.

## 1. INTRODUCTION

The ferroic crystals changes their crystallographic structure when they are subjected to an electric field, or a magnetic field, or a mechanical stress, or a combination of these fields. The crystals that are obtained after applying these field are distinguished by spontaneous magnetization, polarization and strain are called primary ferroic crystals. Also the ferroic crystals undergoes different phase transitions when they are subjected to different temperatures, pressures, and by doping other materials into them. Based on the conditions that are applied to the crystal, phase transitions occur. Based on the resultant phase transition, the physical properties exhibited by the crystal changes.

The physical properties that are exhibited by the crystals after phase transitions can be estimated with the help of symmetry exhibited by the crystals. Using the symmetry of the crystals, their space groups and the corresponding point groups can be identified, with which the physical properties of the crystals can be estimated. The tensors of the crystal physical properties are calculated using the points groups of the crystals before (parent phase) and after (result phase) the phase transition. The spontaneous property tables are constructed using the tensors. These tables helps in estimating the physical properties existence in the crystals. The property tensors for all 1601 Aizu species are calculated by Litvin (2014). The dichromatic matrices of the property coefficients for all 1601 Aizu species are constructed by Litvin and Janovec (2014).

The spontaneous property tables for the crystal (1-x)Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - xPbTiO<sub>3</sub> at three different phase transitions are constructed in this paper to estimate the existence of the 16 physical properties.

## 2. MATERIALS AND METHODS

**Multi-ferroic Phase transitions of PMN-PT:** The spontaneous physical property tables are the tools that helps to identify the existence and changes in 16 physical properties in the single crystal (1-x)Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - xPbTiO<sub>3</sub> (which belongs to the perovskites family) in multi-ferroic phase transition.

According to the phase diagram of the (1-x)Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - xPbTiO<sub>3</sub> system, the crystal have the tetragonal (4mm) ferroelectric phase immediately below the Curie temperature (Bokov, 2005). Two phase transitions exist within the range from room temperature to 250 °C. They are O(orthorhombic) - to - T(tetragonal) and T(tetragonal) - to - C(cubic) phase transition identified as the temperatures 84 °C and 152 °C, respectively (Ye, 2001).

The studies using optical domains showed that the R3m (underline point group is 3m(Bradley, 1972) and P4mm (underline point group is 4mm (Bradley, 1972) phases may coexist at room temperature, with transformation from the R3m into the P4mm phase taking place over a wide temperature interval (Ye, 2000).

The results of the study of (1-x)Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub> - xPbTiO<sub>3</sub> [PMN-35%PT] single crystals using a high-resolution synchrotron X-ray diffraction by Ye(2001) and others showed unambiguous evidence of a monoclinic phase, which is stable between the rhombohedral and tetragonal phases near the morphotropic phase boundry MPB.

Among the phase transitions of the lead magnesium niobate, (1-x)PMN-(x)PT stated above, the phase transitions  $\bar{m}3mF4mm$ ,  $\bar{m}3mF3m$ , and  $\bar{m}3mFmm2$  between the (room temperature) 32 °C to 250 °C when x = 0.3 are considered for current study. The spontaneous physical property tables of the crystal at these phase transitions are constructed.

**List of physical properties:** The following is the list of physical properties calculated for PMN-PT crystal.

**Table.1. List physical property and their Jahn tensor notations.**

Physical Property	Ferroic Type	Jahn tensor notation
Spontaneous strain	Ferroelastic	$[V^2]$
Spontaneous magnetization	Ferromagnetic	$aeV$
Spontaneous polarization	Ferroelectric	$V$
Spontaneous toroidal moment	Ferrotoroidic	$aV$
Elastic compliance	Ferrobielastic	$[[V^2]^2]$
Magnetic susceptibility	Ferrobimagnetic	$[V^2]$
Electric susceptibility	Ferrobielectric	$[V^2]$
Toroidic susceptibility	Ferrotoroidic	$[V^2]$
Piezoelectric coefficient	Ferroelastolectric	$V[V^2]$
Piezomagnetic coefficient	Ferromagnetoelastic	$aeV[V^2]$
Piezotoroidic coefficient	Ferroelastotoroidic	$aV[V^2]$
Magnetolectric coefficient	Ferromagnetolectric	$aeV^2$
Magnetotoroidic coefficient	Ferromagnetotoroidic	$eV^2$
Electrotoroidic coefficient	Ferroelectrotoroidic	$aV^2$

The notations (Jahn 1946) used in the table are as follows:

- $V$  – denotes a polar vector
- $a$  – is a scalar which inverts under time inversion
- $e$  – is a scalar which inverts under spatial inversion
- $[ ]$  – denotes symmetrisation of the enclosed tensor
- $V, aV,$  and  $aeV$  are tensor given as column matrices

**Spontaneous physical property table:** The following table gives the extended matrices of the spontaneous property tables represent a set of equations that relate physical properties of the ferroic materials.

**Table.2. Jahn notation: transformational properties of the physical property tensors**

1	$V$	$aeV$	$[V^2]$	$aV$
$V$	$[V^2]$	$aeV^2$	$V[V^2]$	$aV^2$
$aeV$	$aeV^2$	$[V^2]$	$aeV[V^2]$	$eV^2$
$[V^2]$	$V[V^2]$	$aeV[V^2]$	$[[V^2]^2]$	$aV[V^2]$
$aV$	$aV^2$	$eV^2$	$aV[V^2]$	$[V^2]$

The set of equations that represents the physical properties are given in the extended spontaneous property table (Litvin, 2014). The table gives the transformational properties of the physical property tensors in Jahn (1949) notation.

**Construction of Spontaneous property tables using tensors of the physical property:** The spontaneous property tables are constructed based on the spontaneous coefficients. The spontaneous coefficients are the physical property coefficients that arise in the phase transitions which are zero in its (parent) high symmetry phase and non-zero in low symmetry phase. By comparing the tensors of the physical property in high symmetry with the corresponding physical property in low symmetry, the spontaneous coefficients are identified. Using these coefficients spontaneous property tables are constructed.

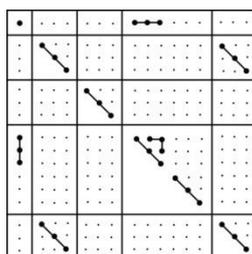
Symbols are used as keys to denote and distinguish the different components. The keys for the phase transition tables are obtained by comparing the tensors of each property in the current phase transition with the corresponding tensors in its parent phase. The properties that exist in both high and low symmetry phases, but changes the way of their presence are given in green color keys. The spontaneous coefficients that are zero in high symmetry phase and non-zero in low symmetry phase are indicated with red color keys.

The description of the keys is given below:

Key	Description
•	- denotes a zero component.
●	- denotes a non-zero component.
●	- (green dot) denotes a non-zero component that is present differently in higher and lower symmetries.
●	- (red dot) denotes a non-zero component that is zero in higher symmetry and non-zero in lower symmetries.
○	- (red circle) denotes a non-zero component with negative sign that is zero in higher symmetry and non-zero in lower symmetries.
●—●	- (two green dot) denotes two equal component that are present differently in higher and lower symmetries.

-  - (two red dot) denotes two equal component that are zero in higher symmetry and non-zero in lower symmetries.
-  - (green dot and circle) denotes two numerically equal but opposite in sign component that are present differently in higher and lower symmetries.
-  - (red dot and circle) denotes two numerically equal but opposite in sign component that are zero in higher symmetry and non-zero in lower symmetries.
-  - (Green square) is equivalent to  $(T_{11}-T_{12})/2$ .

**Spontaneous physical property table of  $m\bar{3}m$**  : The following is the spontaneous physical property table based on the parent point group  $m\bar{3}m$  exhibited by the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ .

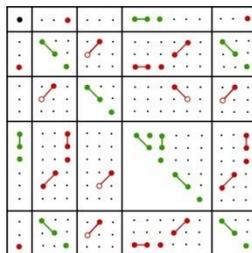


**Figure.1. Spontaneous physical property table of the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$**

The spontaneous property table in the Figure.1 is constructed using the matrices of physical property tensors (Litvin, 2013). From the above table, it can be observed that 6 ferroic properties exists and they are symmetric about the diagonal.

### 3. RESULTS AND DISCUSSION

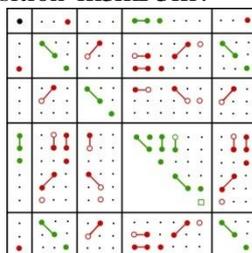
**Spontaneous physical property table of  $m\bar{3}mF4mm$**  : The following is the spontaneous physical property table constructed using the physical property tensors based on the point group  $4mm$  exhibited by the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$  in the phase transition  $m\bar{3}mF4mm$ .



**Figure.2. Spontaneous physical property table of the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$  in the phase transition  $m\bar{3}mF4mm$**

The spontaneous property table in the Figure.2 is constructed using the matrices of physical property tensors (Litvin, 2013) and comparing corresponding cells in the Figure.1

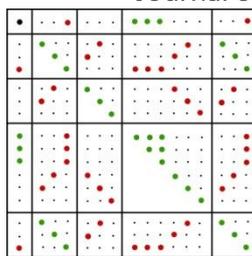
**Spontaneous physical property table of  $m\bar{3}mF3m$**  : The following is the spontaneous physical property table constructed using the physical property tensors based on the point group  $3m$  exhibited by the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$  in the phase transition  $m\bar{3}mF3m$ .



**Figure.3. Spontaneous physical property table of the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$  in the phase transition  $m\bar{3}mF4mm$**

The spontaneous property table in the Figure 4.4.1 is constructed using the matrices of physical property tensors (Litvin, 2013) and comparing corresponding cells in the Figure.1

**Spontaneous property table of  $m\bar{3}mFmm2$**  : The following is the spontaneous physical property table constructed using the physical property tensors based on the point group  $mm2$  exhibited by the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$  in the phase transition  $m\bar{3}mFmm2$ .



**Figure.4. Spontaneous physical property table of the crystal  $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$  in the phase transition  $\bar{m}3mF4mm$**

The spontaneous property table in the Figure 4 is constructed using the matrices of physical property tensors (Litvin, 2013) and comparing corresponding cells in the Figure.1.

From the tables in the Figure2, Figure 3 and Figure 4, it can be observed that all the ferroic properties listed in table .1 are exists and they are symmetric about the diagonal. Along with 6 properties that are exhibited by the crystal in high symmetry, additional properties are exhibited in its lower symmetries after the phase transitions.

#### 4. CONCLUSION

The phase transitions of the crystal PMN-PT changes its cubic structure ( $\bar{m}3m$  (Oh group) prototypic point group) into rhombohedral structure ( $3m$  ( $C_{3v}$ , group) ferroic point group). Also this crystal changes it structure into orthorhombic ( $mm2$  ( $C_{2v}$ , group) ferroic point group) and tetragonal structure ( $4mm$  ( $C_{4v}$ , group) ferroic point group) at different temperatures. The crystal doesn't exhibit magnetic property. It exhibits all the physical properties that are listed in the table1.

In this paper, for each of the ferroic phase transition of the crystal PMN-PT, the physical property tensor matrices with ferroic properties listed in table .1 are constructed. Using these matrices, spontaneous property table are constructed which helps to identify the physical properties exhibited by the crystal in the respective phase transition.

The spontaneous property tables are tools that can be used to read the physical properties exhibited by the crystal. Laborious laboratory work and experimenting which involves high expenditure to identify the physical properties exhibited by the crystals can be avoided by using spontaneous property tables.

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