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## Data Article

# Multiferroic $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ ceramics: Ferroelectricity, ferromagnetism and colossal magneto-capacitance effect

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## ABSTRACT

The mutual control of the electric and magnetic properties of a multiferroic solid is of fundamental and great technological importance. In this article, the synthesis procedure of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics was briefly described and the data acquired for the materials characterization is presented. This data article is related to the research article-Acta Mater. 2016, 121, 144 (<http://dx.doi.org/j.actamat.2016.08.083>). Electric polarization hysteresis loop and I-V curve, which help to confirm the ferroelectricity of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics, were presented. Strong magnetic polarization data was also presented. The great variation of the dielectric constants along with the magnetic field has been presented which helped to demonstrate the giant magnetocapacitance of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ . All the datasets were collected at room temperature. Large ferroelectricity, strong magnetism and colossal magneto-capacitance effect have been all realized in one single phase  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  at room temperature.

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## Specifications Table

Subject area	Physics
More specific subject area	Materials physics, functional materials.
Type of data	figures
How data was acquired	Instruments used- Bruker D8 Advance for XRD measurement, ZT-IA ferroelectric measurement system, Quantum Design physical property measurement system (PPMS), Wayne Kerr 6500B LCR station.
Data format	Raw, analyzed, etc
Experimental factors	The ceramics were heat treated in oxygen atmosphere at 700 °C for three times and then both surface sides were coated with silver electrode for ferroelectric measurement and magnetic field-dependent capacitance measurement. The powders for magnetic characterization measurement have also been annealed in O <sub>2</sub> for three times.
Experimental features	The magnetocapacitance data was measured by placing the specimen in a space between two magnets. The magnetic field was adjusted by voltage and current of the coils of the magnets. The measurement was performed under different frequencies at room temperature.
Data source location	Wuhan University of Technology, Wuhan, China,
Data accessibility	Data is supplied with in this article.

## Value of the data

1. Electric polarization data was used to confirm the ferroelectricity of the La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> ceramics at room temperature.
2. Magnetic polarization data was used to demonstrate the ferromagnetism of the La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> at room temperature.
3. The variable dielectric constants upon magnetic field were used to display the giant magnetocapacitance effect of the La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> ceramics at room temperature.
4. The three datasets demonstrate not only the coexistence of large ferroelectricity and strong ferromagnetism in single La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> at room temperature, but also the realization of its strong magnetoelectric coupling effect at room temperature.

## 1. Data

The data given in this data article is in the form of three figures. It describes briefly the preparation process of the La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> ceramics, the electric and magnetic characterization of the La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> ceramics as well as its magnetoelectric coupling phenomenon. The data article includes ferroelectric and I–V datasets giving the information of the electric polarization and two current peaks upon applied field, ferromagnetic datasets and variable magnetic field dependent-capacitance datasets.

## 2. Experimental design, materials and methods

The single-phase La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> powders were prepared by a polymer precursor method, certain amount of power was pressed into a pellet which was sintered into ceramic. After sintering, the ceramic has subsequently experienced heat-treatment in O<sub>2</sub> at 700 °C three times to remove the oxygen vacancies and transform Fe<sup>2+</sup> into Fe<sup>3+</sup> in the La<sub>0.2</sub>Pb<sub>0.7</sub>Fe<sub>12</sub>O<sub>19</sub> ceramic. for P-E hysteresis loop measurement, both sides of the ceramic surfaces were coated with silver paste as electrode

which was sintered at 820 °C for 15 min. Then the ferroelectric hysteresis loop was measured upon the polycrystalline  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  pellet with electrodes by using an instrument referred as ZT-IA ferroelectric measurement system. Magnetization measurement was carried out on a Quantum Design Physical Property Measurement System (PPMS). The magnetocapacitance parameters of the  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics were measured using a Wayne Kerr 6500B LCR station or an IM 3533-01 LCR meter by applying a variable magnetic field on the specimen.

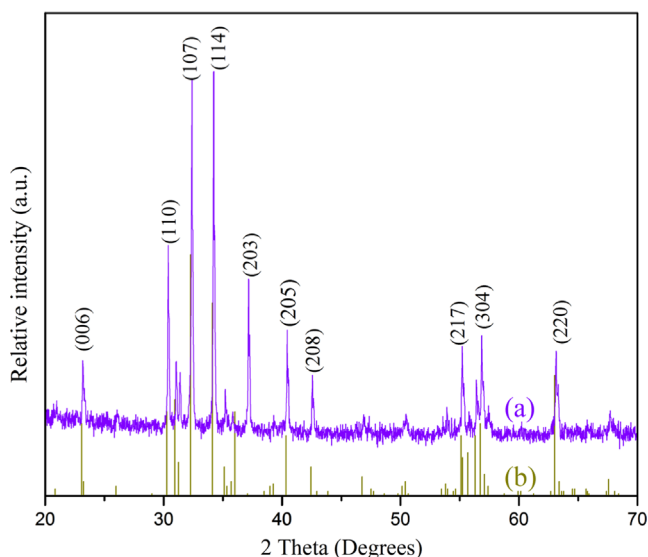
### 3. Multiferroic characterization of $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ ceramics

#### 3.1. Structure identification

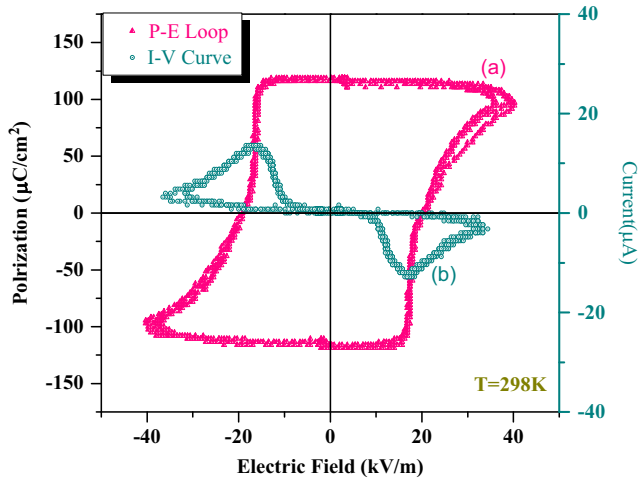
Fig. 1a shows XRD pattern of  $\text{O}_2$  treated  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics; the underneath red diffraction lines are coming from the standard diffraction spectrum of  $\text{PbFe}_{12}\text{O}_{19}$  (PDF#15–0623). The CIF file from Rietveld refinement has been included in this article. The  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics with oxygen treatment is identified to be magnetoplumbite-5H structure, which is the same structure of  $\text{PbFe}_{12}\text{O}_{19}$ . The Rietveld refinement results suggest that La atoms have been successfully replaced Pb atoms in  $\text{PbFe}_{12}\text{O}_{19}$  and has entered into its crystal lattice. The lattice unit has been contracted about 0.44% after the replacement of Pb atoms with La atoms. The overlap of the two diffraction patterns reveals that  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  shares the same crystal structure with  $\text{PbFe}_{12}\text{O}_{19}$ .

#### 3.2. Ferroelectricity and $I$ – $V$ features

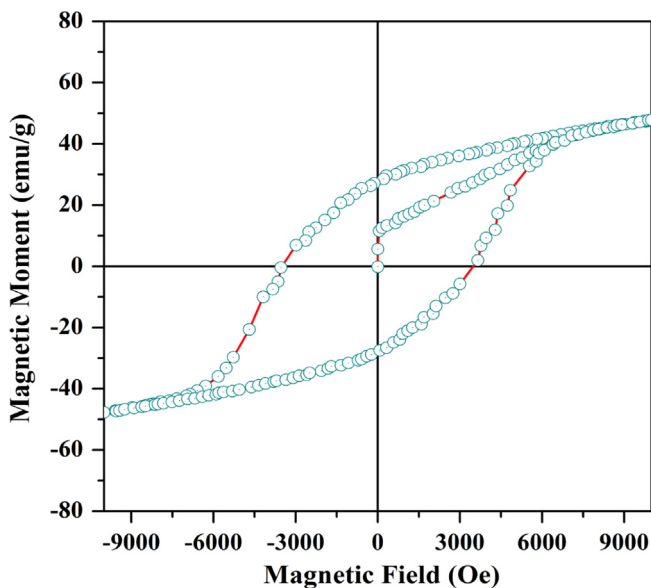
Fig. 2a displays a electronic polarization hysteresis loop of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics with full saturation. The ferroelectric properties were measured upon a lab-constructed ZT-I system at a frequency of 33 Hz and 298 K. The polarization loop looks like that of most standard ferroelectric compounds. The remnant polarization of the hysteresis loop is about  $119.4 \mu\text{C}/\text{cm}^2$ . This value is a little higher than that of pure  $\text{PbFe}_{12}\text{O}_{19}$  ceramic ( $104 \mu\text{C}/\text{cm}^2$ ) [1] obtained by the same process. The coercive field of the  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics is determined to be 20.6 kV/m.



**Fig. 1.** Structure identification of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ . (a) XRD pattern of the  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  with  $\text{O}_2$  annealing process. (b) The standard diffraction pattern of the  $\text{PbFe}_{12}\text{O}_{19}$  compound (PDF#15–0623) being marked by red discrete lines. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

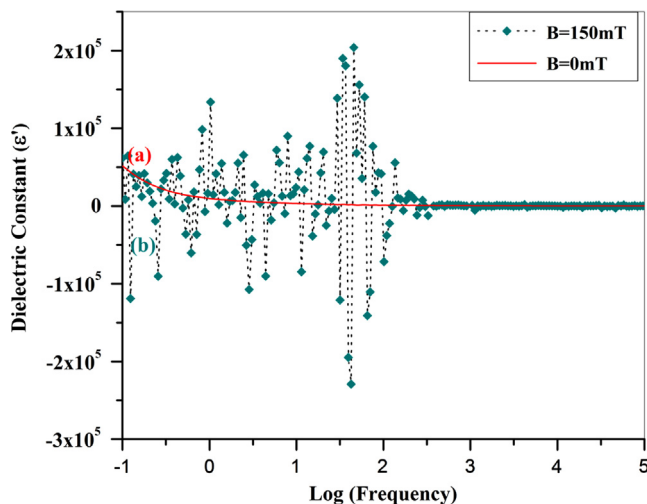


**Fig. 2.** Ferroelectric characterization of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ : (a) The ferroelectric hysteresis loop of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramic being measured at a frequency of 33 Hz and 298 K; (b) the nonlinear I–V curve of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramic. The ceramic was sintered at 1000 °C for 1 h and subsequently heat-treated in an  $\text{O}_2$  atmosphere for 9 h. The measurement was performed at room temperature.



**Fig. 3.** Magnetic hysteresis loop of polycrystalline  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramic being sintered at 1000 °C for 1 h and subsequently annealed in  $\text{O}_2$  for 9 h. The measurement was performed at room temperature.

Fig. 2b demonstrates two I–V peaks at the vicinity of positive and negative coercive field. The two particular peaks present a great nonlinear relationship between voltage (V) and current (I), which provides us with an additional evidence for the ferroelectricity of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics and could exclude the possibility of current leakage. The classic ferroelectric hysteresis loop with full saturation and two nonlinear I–V peaks convinced us that  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  is indeed a ferroelectric



**Fig. 4.** Frequency-dependent dielectric constant of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics upon the magnetic field of (a)  $B=0$  mT (red solid line) and (b)  $B=150$  mT (dark cyan dotted line). The specimen was air-sintered at  $1000^\circ\text{C}$  for 1 h and subsequently annealing in  $\text{O}_2$  for 9 h. The measurement was performed at room temperature. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

compound. The origin of the ferroelectricity of M-type lead hexaferrite has been discussed in detail in the previous literatures [1,2].

### 3.3. Ferromagnetism

Fig. 3 demonstrates the magnetization hysteresis loop of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ . The remnant magnetic moment ( $M_r$ ) is about 28.7 emu/g and the coercive magnetic field ( $H_c$ ) is around 3439.2 Oe. The remnant moment of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  has been reduced by a small amount while its coercive field has been enhanced by 1114 Oe in comparison with that of pure  $\text{PbFe}_{12}\text{O}_{19}$  [1], since the substituted La provides additional unpaired f electrons, which create more magnetization moment. The large hysteresis loop reflects the strong magnetic feature of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$ .

### 3.4. Giant magnetocapacitance effect

In order to check out the magnetoelectric coupling effect of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics, we then measured the capacitance of the specimens at different frequencies upon a applied magnetic field. The specimen was coated with silver electrodes on both surfaces and then put in a space between two magnets, the frequency-dependent capacity data was measured by a IM3533-1 LCR Meter (Hioki Company, Japan) without and with applying a magnetic field at  $B=150$  mT. Fig. 4b shows the great response of capacity (or dielectric constant) to the magnetic field as a function of frequencies for  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  ceramics. We found that there was a giant response of the capacitance (dielectric constant) to the magnetic field at low frequency region. There is big difference between the dielectric constants without (Fig. 4a) and with (Fig. 4b) external magnetic field. Upon the applied magnetic field ( $B=150$  mT), the dielectric constant ( $\varepsilon(B)$ ) could oscillate along with  $\varepsilon(0)$  in a great amplitude, varying from 204065.6 to  $-229257.9$ . The dielectric constant ( $\varepsilon(B=150$  mT)) was determined to be 68184 at the frequency of 50 Hz and 42951 at 100 Hz, respectively (Fig. 4b); while the corresponding dielectric constants at  $B=0$  ( $\varepsilon(0)$ ) equal to 921.4 at 50 Hz and 731.6 at 100 Hz, respectively. The maximum value of the magnetocapacitance ratio exceeds  $2.18 \times 10^4\%$  ( $\Delta\varepsilon(B) = (\varepsilon(B) - \varepsilon(0)) / \varepsilon(0)$ ,  $B=150$  mT and  $f=50$  Hz). These values match well with that being measured by a Wayne Kerr 6500B LCR station [3]. The agreement between two sets of capacitance data ( $B=150$  mT) being measured

from two different LCR meters indicates the reliability and repetitiveness of the B-field dependent dielectric constants.

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### **Transparency document. Supporting information**

Transparency data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.dib.2016.11.067>.

### **Appendix A. Supporting information**

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.dib.2016.11.067>.

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- [3] G. L. Tan and H. H. Sheng, Multiferroism and Colossal Magneto-Capacitance Effect of  $\text{La}_{0.2}\text{Pb}_{0.7}\text{Fe}_{12}\text{O}_{19}$  Ceramics. *Acta Mater.* 121, 2016, 144.