# Investigation and Improvement of the Physical Properties of Multiferroic with Formula La<sub>1-x</sub>Sb<sub>x</sub>FeO<sub>3</sub>

#### **Thesis**

Submitted for the Partial Fulfillment of the Degree of M.Sc. of Physics

By

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#### Approval Sheet

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## **DEDICATED**

To

My **Father** 

My **Mother** 

My **Brothers** 

My **Sister** 

And

My **Doctors** 

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#### **Abstract**

It is well know that Lanthanum orthoferrite LaFeO<sub>3</sub> has an antiferromagnetic behavior and BiFeO<sub>3</sub> is a typical multiferroic. Therefore, we expected that doping LaFeO<sub>3</sub> by antimony (Sb) which lies in the same group with Bi in the periodic table gives good electric properties for LaFeO<sub>3</sub> keeping high magnetic properties. Consequently, we will achieve multiferroic behavior.

The experimental steps in the thesis are summarized into the following: preparation and characterization of La orthoferrite system. Improvement of ferroelectric like behavior of the orthoferrite under investigation by the partial replacement of  $\mathrm{Sb}^{3+}$  instead of  $\mathrm{La}^{3+}$ .

A correlation between structural, magnetic and electric properties will be established to outline the enhancement of the properties of the prepared samples. Examination of the multiferroic behavior of the samples will open a new era of many applications.

The techniques and characterization methods used in this study are: Structural analysis using X-ray diffraction (XRD), Spectroscopic analysis using Fourier transform infra red (FTIR) and transmission electron microscope (TEM). Magnetic properties by measuring magnetic susceptibility using the Faraday's methods were measured. Magnetic hysteresis at different temperatures and magnetic field intensities and the electrical properties by measuring dielectric constant, dielectric loss, ac conductivity, Seebeck voltage coefficient was also measured.

The thesis consists of two groups. The effect of the substitution of  $Sb^{+3}$  ions instead of  $La^{+3}$  ions was studied in the first group of the formula  $La_{1-x}Sb_xFeO_3$  ( $0 \le x \le 0.3$ , step 0.05) where the parent is  $LaFeO_3$ . From XRD the samples with  $Sb^{+3}$  content (x) 0.0, 0.05 and 0.1 were formed in single phase, but the samples with x > 0.1 have a small secondary phase from  $Sb_2O_3$ .

The shapes of the particles and the crystallite size were studied by using TEM. The particles have different shapes such as rods and dots which caused a great improvement in the measured physical properties of the samples. IR was used to assure the formation of the samples in the proper form.

There is an enhancement in the magnetic properties by adding  $Sb^{+3}$  ions especially in small amount i.e. x=0.05 where the molar magnetic susceptibility ( $\chi_M$ ) increases from 0.00916 emu/gm.mole for the  $LaFeO_3$  to 0.0269 emu/gm.mole for the  $La_{0.95}Sb_{0.05}FeO_3$  at temperature (T) = 500 K and magnetic field intensity 3050 Oe. The magnetic hyteresis was used to calculate the magnetic constants such as coersive field ( $H_C$ ), remenance magnetization ( $M_r$ ) and saturation magnetization ( $M_s$ ). The  $H_C$  increased from 1196 Oe for the  $LaFeO_3$  to 6666 Oe for the sample  $La_{0.95}Sb_{0.05}FeO_3$ . The value of  $H_C$  of the sample containing  $Sb^{+3}$  content 0.05 was increased 6 times than that of the parent  $LaFeO_3$ . Ms increases from 0.1614 emu/gm for the parent  $LaFeO_3$  to 0.2654 emu/gm for the  $La_{0.95}Sb_{0.05}FeO_3$ .

Not only the magnetic properties were enhanced but also, the electrical properties were improved. The conductivity ( $\sigma$ ) increases from 0.002366 Ohm<sup>-1</sup>.m<sup>-1</sup> for the LaFeO<sub>3</sub> to 0.0301 Ohm<sup>-1</sup>.m<sup>-1</sup> for the sample La<sub>0.95</sub>Sb<sub>0.05</sub>FeO<sub>3</sub> at T = 553 K and frequency 1MHz. Also, the dielectric constant ( $\epsilon$ ) was increased with increasing the Sb<sup>+3</sup> content.

The sample  $La_{0.95}Sb_{0.05}FeO_3$  has antiferromagnetic and ferroelectric properties so, it has a multiferroic behavior.

In the second group, we studied the substitution of divalent ions  $(Ca^{+2} \text{ and } Sr^{+2})$  on the expense of  $La^{+3}$  ions. The XRD showed that the samples were formed in a single phase. TEM was used to show the shapes of the particles.

There is an enhancement in the electrical properties where  $\epsilon'$  increased from 74.15 for the LaFeO<sub>3</sub> to 9623.06 for the sample La<sub>0.7</sub>Ca<sub>0.3</sub>FeO<sub>3</sub> at T = 500 K and frequency

1MHz. Also, the conductivity ( $\sigma$ ) increases from 0.000831 Ohm<sup>-1</sup>.m<sup>-1</sup> for the LaFeO<sub>3</sub> to 0.48388Ohm<sup>-1</sup>.m<sup>-1</sup> for the sample La<sub>0.7</sub>Ca<sub>0.3</sub>FeO<sub>3</sub> at T = 500 K and frequency 1MHz.

There are changes in the magnetic properties of the parent sample by the substitution of the divalent ions. The decrease in the Néel temperature was observed from 763 K of  $LaFeO_3$  to 748 K of  $La_{0.7}Sr_{0.3}FeO_3$  and 694 K for  $La_{0.7}Ca_{0.3}FeO_3$ .

The improvement in the physical properties of the LaFeO<sub>3</sub> using substitution by either trivalent ion  $Sb^{+3}$  ( as in first group ) or divalent ions  $Ca^{+2}$  and  $Sr^{+2}$  ( as in second group ) instead of La<sup>+3</sup> is a good chance to use the prepared samples in many applications.

#### Aim of the Work

- -Preparation and characterization of La orthoferrite system by double ceramic technique.
- -Improvement of ferroelectric like behavior of the orthoferrite under investigation by the partial substitution of the Sb<sup>3+</sup> ions in stead of La<sup>3+</sup> ions.
- -Enhancement of the magnetic interaction of the samples.
- -Examination of the multiferroic behavior of the prepared samples.

Table(5.1) Illustrates IR for the samples of the first group  $La_{1-x}Sb_xFeO_3;\ 0.0 \le x \le 0.3.$ 

X	$v_1(\text{cm}^{-1})$	$v_2(\text{cm}^{-1})$	$v_3$ (cm <sup>-1</sup> )	υ <sub>4</sub> (cm <sup>-1</sup> )	υ <sub>5</sub> (cm <sup>-1</sup> )	$v_6(\text{cm}^{-1})$
0.0	980.0	613.3	543.3	363.3	293.3	225.0
0.05	976.7	616.7	541.7	350.0	295.0	220.0
0.1	981.7	616.0	541.7	353.0	295.0	218.3
0.15	983.3	623.3	543.3	346.7	300.0	223.3
0.2		620.0	543.3	360.0	313.3	221.5
0.25		606.7	546.7	360.0	293.3	220.0
0.3	985.0	610.0	545.0	363.3	303.0	

Table (5.2) The values of the Curie constant, the Curie – Weiss constant and the effective magnetic moment of the samples  $La_{1-x}Sb_xFeO_3$ ;  $0.0 \le x \le 0.3$  at the magnetic field intensity 2620 Oe.

X	θ (Κ)	C (emu/gm.mole).K	$\mu_{effe}$ (B.M.)
0.0	-170	3.96	5.6
0.05	-165	18.2	12.07
0.1	-160	7.09	7.54
0.15	-110	12.3	9.93
0.20	-123	7.52	7.76
0.25	-165	2.93	4.84
0.30	-160	6.63	7.28

Table (5.3) Dependence of the activation energies ( $E_1$  at low temperature and  $E_2$  at high temperature) on the frequency.

Frequency (MHz)	E <sub>1</sub> (ev)	E <sub>2</sub> (ev)
1	0.40	1.36
1.2	0.38	1.36
1.5	0.36	1.36
2	0.31	1.36
2.5	0.23	1.36
3	0.35	1.36
4	0.32	1.36
5	0.25	1.36