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Magnetoelectric properties of $0.1Bi_{0.95}Dy_{0.05}FeO_3$-$0.9Pb(Fe_{2/3}W_{1/3})O_3$ multiferroic

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Abstract. Pb-based complex perovskites with Fe^{3+} like $Pb(Fe_{2/3}W_{1/3})O_3$ (PFW) were found to be interesting because of their unique properties such as relaxor and magnetoelectric behavior. PFW is ferroelectric with ferroelectric Curie temperature T_C between 150 and 200 K and at the same time is antiferromagnetic with magnetic Neel temperature about 400 K. $BiFeO_3$ is a well known perovskite compound which exhibits ferroelectric (T_C = 1103 K) and antiferromagnetic (T_N = 643 K) ordering simultaneously. The polycrystalline sample of $0.1Bi_{0.95}Dy_{0.05}FeO_3$ – $0.9Pb(Fe_{2/3}W_{1/3})O_3$ were synthesized using standard sintering procedure. Magnetization vs. magnetic field (at 4.2 K) curves was measured. Magnetoelectric properties of the sample were obtained.

1. Introduction

Materials that exhibit ferromagnetic and ferroelectric orderings in the same phase are named multiferroics. Magnetoelectric materials are very attractive due to a strong coupling between magnetic and electric subsystems, what means that magnetic field is able to induce electric polarization and vice versa. Magnetoelectrics are quite interesting because of their fundamental aspects in modern physics but also they are very desirable for practical applications. The first single phased multiferroic perovskites were discovered in the early 1960s. However, very rare and limited progress has been made during the last several decades [1-6]. A significant development of multiferroic materials has been started with successful synthesis of multiferroics' thin films [7].

Unfortunately, most of the multiferroic materials is characterized by a low magnetic ordering temperature, considerably below room temperature, and therefore a magnetoelectric effect is relatively small. Bismuth ferrite $BiFeO_3$ is a well known perovskite compound which simultaneously exhibits at ambient temperature ferroelectric (T_C = 1110 K) and antiferromagnetic (T_N = 610 K) ordering. Stereochemical activity of the Bi lone-pair electrons induces ferroelectric polarization, while a

partially field 3d orbitals of the Fe^{3+} ions produce G-type antiferromagnetic ordering. $BiFeO_3$ has a spatially modulated magnetic structure of cycloidal type with the period of modulation about 62 nm [4]. One of the way to suppress the spiral spin modulation is a chemical substitution of the magnetically active atoms, especially rare earth trivalent ions into Bi sublattice.

$Pb(Fe_{2/3}W_{1/3})O_3$ is a multiferroic in which magnetically active Fe^{3+} cations are partailly substituted by ferroelectrically active W^{6+} cations. This compound is characterized by the ferroelectric Curie temperature between 150 and 200K and at the same time is antiferromagnetic with magnetic Neel temperature about 400 K [8].

Unfortunately, high leakage current, secondary oxide phases and other defects make $BiFeO_3$ difficult to obtain [2,3,4]. Therefore, solid solution of Dy-doped $BiFeO_3$ and $Pb(Fe_{2/3}W_{1/3})O_3$ was synthesized. In this paper, we report synthesis, crystal structure and magnetoelectric properties of $0.1Bi_{0.95}Dy_{0.05}FeO_3$-$0.9Pb(Fe_{2/3}W_{1/3})O_3$.

2. Experimental

High purity powder oxides of Bi_2O_3, Dy_2O_3, Fe_2O_3, PbO_2 and Fe_2WO_3 were weighted in stoichiometric proportions and mechanically activated for 7h during a ball milling process. Thereafter, mixture was calcined at 780°C for 4h, secondly granulated and pressed into disc shaped pellet (1.8mm height and 1.53cm in diagonal). Finally, pellet was heat-treated at 545°C for 2h and sintered at 800°C also for 2h.

3. Magnetic and magetoelectric properties.

Magnetic hysteresis loop M(H) was recorded at 4.2 K and is presented in Figure 1. The magnetization curve M(H) reveal a typical antiferromagnetic behavior with no magnetic saturation effect up to maximal magnetic field of 56 kOe. More detailed analysis of M(H) curve is showing a slightly ferromagnetic behavior. Evaluated values of remanent magnetization M_R and coercive field H_C are 220.4 emu/mol and 1060 Oe, respectively.

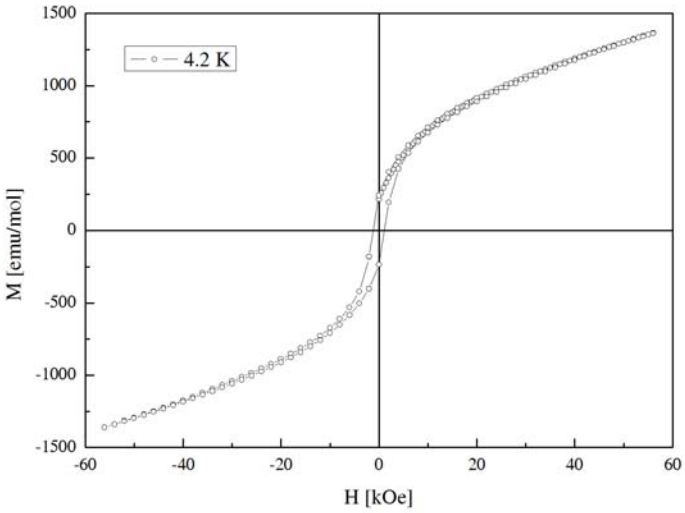

Figure 1. Magnetization curves M(H) taken at 4.2 K.

In order to measure a magnetoelectric voltage coefficient the disc shaped sinter of 0.19 cm height and 1.5 cm in diameter was placed into a static magnetic field H_{DC}, created by electromagnet with additional modulation of magnetic field $H_{AC}(f)$ produced by Helmholtz coils. Both magnetic fields were oriented perpendicular to the surface of sinter. An electric signal $\delta U(f)$ from the sample was detected by using lock-in amplifier. The voltage δU was measured for different magnitudes of static magnetic field and frequencies of the modulation field in the range 200 Hz – 12 kHz. A magnetoelectric voltage coefficient (ME) was derived using formula:

$$ME = \delta U / (\delta H \cdot d) \quad (1)$$

where *d* is a height of the investigated sample.

Figure 2 presents dependence of the ME coefficient in function of the H_{AC} frequency at static magnetic field H_{DC} of magnitude 350 Oe. At low values of frequency the ME coefficient rises rapidly, while with increasing the frequency a monotonically growth of the ME is observed up to about 7 kHz. Further increasing of the H_{ac} field frequency cause slower increase of the ME coefficient up to 12 kHz, which was the maximal value for our experimental set-up.

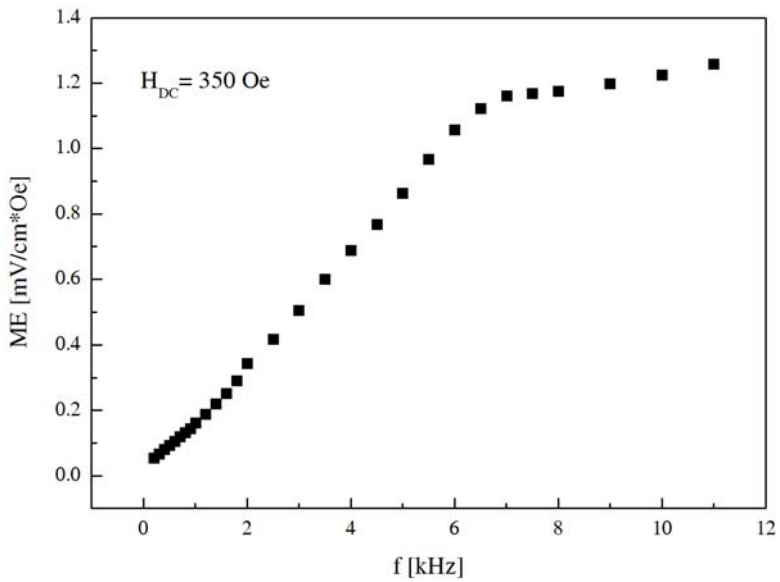

Figure 2. Magnetoelectric voltage coefficient (ME) vs. frequency for $0.1Bi_{0.95}Dy_{0.05}FeO_3$-$0.9Pb(Fe_{2/3}W_{1/3})O_3$.

Figure 3 is showing the ME voltage coefficient vs. static magnetic field for modulated frequency f = 5 kHz and H_{AC} = 4 Oe. Growing the H_{DC} field causes decreasing of the ME parameter up to about 0.5 kOe. Further increasing the field results in increasing of the ME coefficient and the maximal value of about 0.837 mV/cmOe is obtained for the H_{DC} field of about 1 kOe. Increasing H_{DC} field cause rapid decreasing of the ME coefficient at the H_{DC} value of about 2 kOe this decreasing slows down.

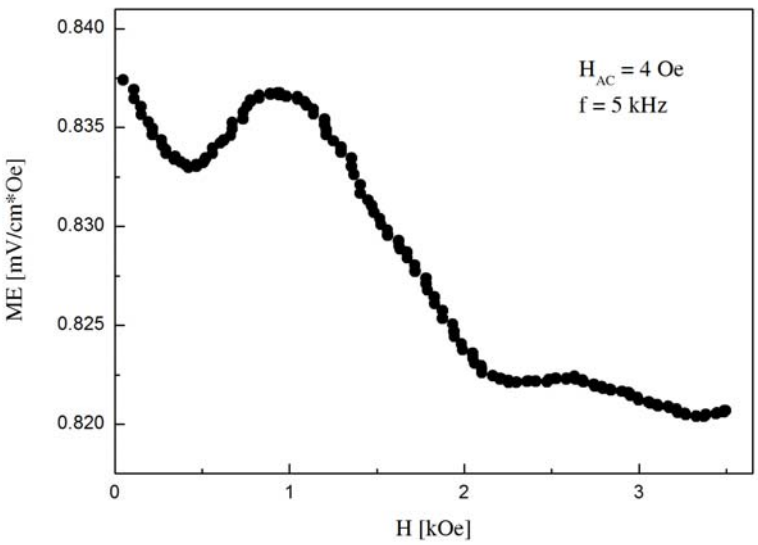

Figure 3. Frequency dependence of the magnetoelectric coefficient.

4. Conclusions

$0.1Bi_{0.95}Dy_{0.05}FeO_3$-$0.9Pb(Fe_{2/3}W_{1/3})O_3$ perovskite was synthesised by means of the conventional solid state reaction method. The magnetic hysteresis loop evidenced the antiferromagnetic behavior of the investigated material with slight ferromagnetic behavior with the remanent magnetization M_R and coercive field H_C are 220.4 emu/mol and 1060 Oe, respectively. The magnetoelectric measurement evidenced the magnetoelectric properties of the obtained solid solution. There was observed strong dependence of the ME coefficient vs. frequency of the H_{AC} field. The complicated $ME(H_{DC})$ dependence was observed.

The obtained results confirmed the magnetoelectric properties of the $0.1Bi_{0.95}Dy_{0.05}FeO_3$-$0.9Pb(Fe_{2/3}W_{1/3})O_3$ solid solution.

Acknowledgements

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