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Multiferroics application - Magnetic controlled piezoelectric transformer[#]

Lucjan Kozielski[1,*], Frank Clemens[2]

[1]*University of Silesia, Department of Materials Science, Sosnowiec, Poland*
[2]*EMPA, Swiss Federal Laboratories for Materials Testing and Research, Laboratory for High Performance Ceramics, 8600 Duebendorf, Switzerland*

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Abstract

Dense lead zirconate titanate (PZT) ceramics is typically used for fabrications of high power piezoelectric devices. In case of lanthanum and iron ions doping into PZT solid solution (PLFZT), material exhibiting both piezoelectric and magnetic properties can be obtained. Among many investigated compositions particularly the $Pb_{0.91}(La_{0.5}Fe_{0.5})_{0.09}(Zr_{0,65}Ti_{0,35})_{0,9775}O_3$, *located near the morphotropic boundary, exhibits the highest magnetoelectric effect. This coupling between magnetization and polarization is achieved by the* Fe^{3+} *ions addition that sufficiently rise sensitivity to magnetic field without decreasing the dielectric loss coefficient at the same time. Taking advantageof this specific material the piezoelectric transformer (PT) with magnetic feedback was fabricated, which converts an electrical AC input voltage into ultrasonic vibrations and reconverts back to an output as AC voltage proportionally to the magnetic field intensity. In the present study the unipoled radial mode piezoelectric transformers based on PLFZT-type ceramics prepared by hot-press sintering have been investigated. The effect of the magnetic field on the operating properties was measured for piezoelectric transformer operating at the first resonance frequency.*

Keywords: *multiferroics, lead zirconate titanate, doping, piezoelectric transformer*

I. Introduction

Piezoelectric resonant based sensors have been investigated by many researchers and different kinds of sensors and biosensors have been reported. A pressure, chemical and temperature sensors have been proposed, as well as an accelerometer sensors based on resonators have been constructed [1]. The use of this method is based on detecting a reflection signal from the resonator against an interrogating signal [2,3]. On the other hand, to the best of our knowledge, there has been only one piezoelectric transformer (PT) based magnetic sensor reported in literature [4], where the authors have taken advantage of ferromagnetic and ferroelectric gradient disk structure. Therefore, we have gone one step further and proposed a multifferoic material monolithic resonator, which consists of only one compact transducer from lanthanum and iron doped lead zirconate titanate (PLFZT) ceramics. Furthermore, to achieve higher sensitivity this device is able to be used as an element in the active sensor matrix.

The latest trend for substitution of a magnetic transformer with piezoelectric transformer has made great progress along with both miniaturization and integration of modern power electronic system. Piezoelectric transformer overcomes traditional largely used electromagnetic type with smaller size, non-electromagnetic radiation, simpler structure and higher conversion efficiency properties. The structure, size, vibration mode and power output method of piezoelectric transformers will affect their own input and output impedance characteristics, thus affecting the output power and efficiency of power conversion. A radial vibration-type piezoelectric transformer is perceived as the most prospective construction. Compared with the other types of transformers, both the output power and conversion efficien-

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[*] Corresponding author: tel: +48 32 3689 564
fax: +48 32 3689 563, e-mail: *lucjan.kozielski@us.edu.pl*

cy of a radial vibration-type piezoelectric transformer are increased significantly in recent times, meanwhile, its voltage transfer ratio is easy to adjust and can meet the requirements of both step-down and step-up applications. A radial vibration-type piezoelectric transformer has a broad range of applications, so it can be effectively used for AC/DC and DC/AC converters, adapters and electronic ballasts [5,6].

Additionally, among many different work patterns and shapes of piezoelectric transformers, the radial vibration disk type is the most sensitive to the influence of foreign factors, so that this particular construction have been chosen to our experiment with magnetic field.

A coupling effect between ferroelectric and ferromagnetic properties is strongly desired for a large number of devices that need effective conversion between magnetic and electric signals. In multiferroic materials, spontaneous electric polarization can be changed by an applied magnetic field and the spontaneous magnetization can be modulated by an external electric field. In multiferroics, the magnitude of such effects is strongly dependent on the efficiency of the elastic coupling between magnetostrictive and piezoelectric components. The exact nature of the magnetoelectric (ME) coupling is not well understood [7], however, a variety of dynamics studies including electron-phonon interactions were published and explained in relevant electronic models [8]. When the neutral phase is magneto excited, the linear behaviour is observed by plotting the interband electronic transitions as a function of the increment in the spin-reorientation transitions. The final efficiency is determined merely by magnetoelectric coupling factor [9]. In order to elucidate and quantify the nature of the ME coupling, it is important to measure the ME coefficient, which can be defined as the ratio of the induced electric field E caused by the applied magnetic field H [10].

Among such materials lead iron niobate (PFN) and lead zirconate titanate (PZT) based ceramics have simple and well known fabrication route but the Fe^{3+} doped PLZT 9/65/35 ceramics appeared as the most sensitive to magnetic field material due to very high values of piezoelectric coefficients together with relatively high ME coupling [11]. Fe^{3+} donor doping in perovskite structure is believed to play a critical role in the magnetic field induced ordering changes, similarly as in PFN ceramics. Therefore this dopant was chosen to our experiment [12].

The main aim of our work is to analyse the influence of magnetic field on electric properties of piezoelectric transformer fabricated from $Pb_{0,91}(La_{1-x}Fe_x)_{0,09}(Zr_{0,65}Ti_{0,35})_{0,9775}O_3$ ceramics for various Fe to La ions ratio.

II. Experimental - Sample preparation

$Pb_{0.91}(La_{1-x}Fe_x)_{0.09}(Zr_{0,65}Ti_{0,35})_{0,9775}O_3$ (PLFZT) ceramics, with $x = 0$, 0.3, 0.5 and 0.8, were prepared by mixed oxide method. A flow chart, presented in Fig. 1, schematically illustrates respective steps of the PLFZT ceramics preparation. The Pb, La, Fe, Zr and Ti oxides were put into poliamid jar with YTZ balls (ZrO_2-Y_2O_3) of 10 mm diameter in ethanol solution. The milling process was carried out in planetary mill for 24 h (200 rpm) in air atmosphere. After that, the obtained powders were dried in air, pressed in steel die into pellets and subjected to thermal processing (solid state synthesis) at 925 °C for 3 hours. Finally, the reacted material was crushed in agate mortar with pestle and shaped into disks with the diameter of 10 mm and 1 mm in height, applying again the pressure of 200 MPa. Final sintering was made using hot pressing (HP) method at 1200 °C with pressure of 20 MPa for 2 hours. The HP was implemented in sample preparation, because this technique was supposed to increase the magnetoelectric (ME) effect for one order of magnitude [10].

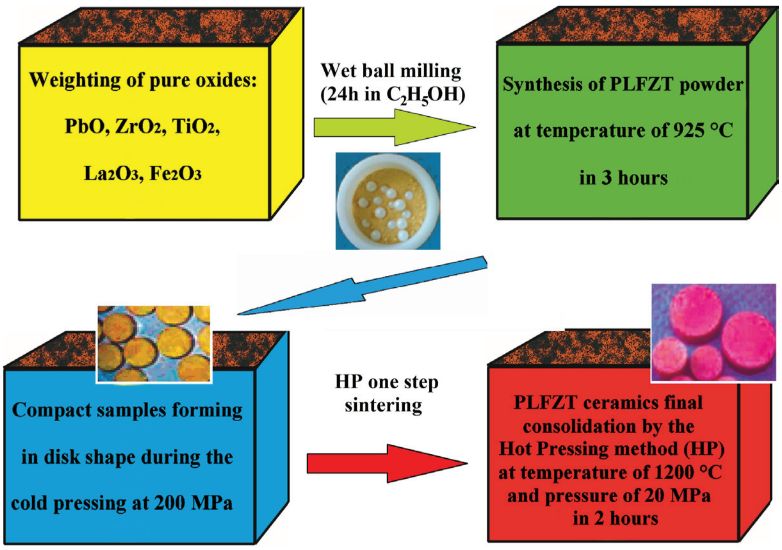


Figure 1. Flow chart of the PLFZT ceramics preparation

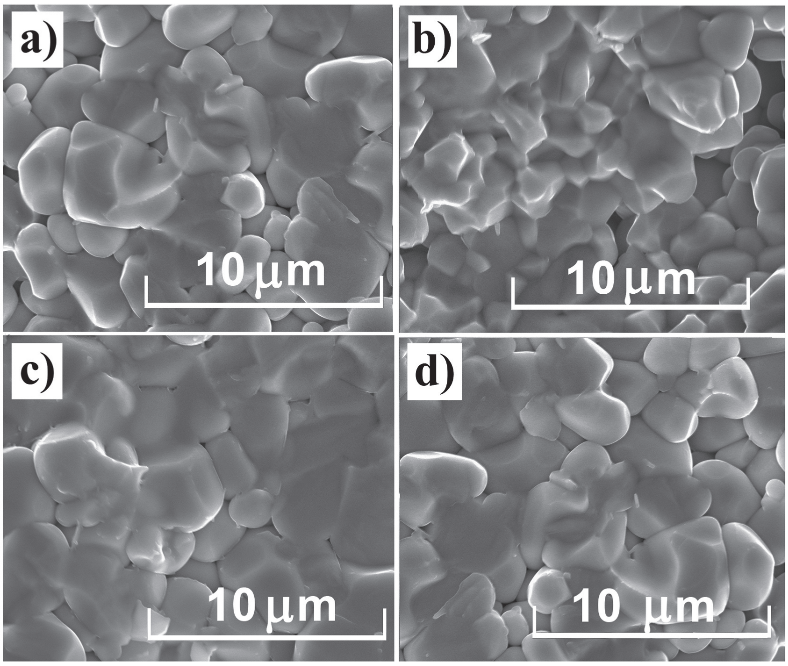


Figure 2. Surface SEM images of $Pb_{0.91}(La_{1-x}Fe_x)_{0.09}(Zr_{0.65}Ti_{0.35})_{0.9775}O_3$ ceramics with: a) x = 0, b) x = 0.3, c) x = 0.5 and d) x = 0.8

III. Results and discussion

3.1 Structural characterization of PLFZT samples

The density (measured by Archimedes method) of the obtained PLFZT ceramics decreased from 7.30 to 7.26 g/cm^3 when Fe^{3+} substitution increases from x = 0 to x = 0.8. The microstructure and grain size of the PLFZT ceramics were investigated using scanning electron microscopy (SEM, HITACHI S-4700). The SEM micrographs of the PLFZT material, is presented in Fig. 2. Grain sizes were found to be uniformly distributed across the samples surface. The average grain size of the PLZT ceramics is equal 2.3 μm (Fig. 2a), although for higher iron content the size of the grains gradually increased to achieve the average grain size of 2.7 μm for the PLFZT ceramics with x = 0.8 (Fig. 2d).

The X-ray spectra of the investigated materials were obtained by PANalytical X'Pert Pro Diffractometer and later analyzed by the program X'Pert HighScore Plus. The recorded diffraction patterns, presented in Fig. 3, are characteristic for pseudo regular perovskites struc-

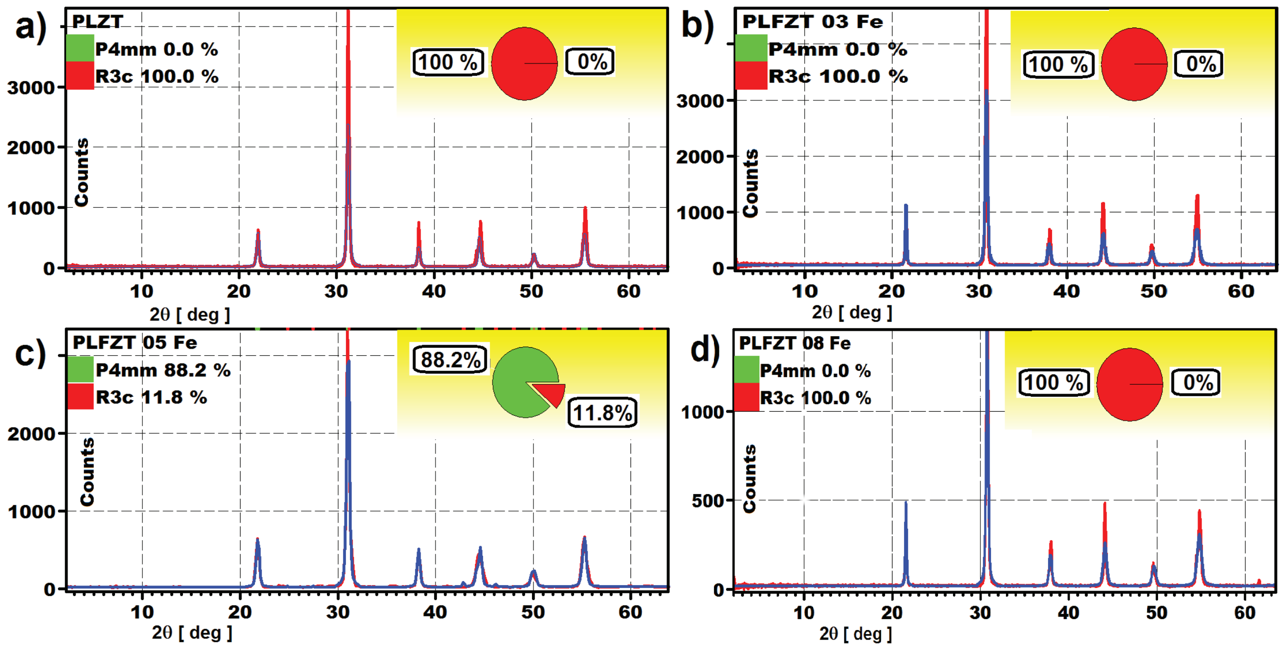
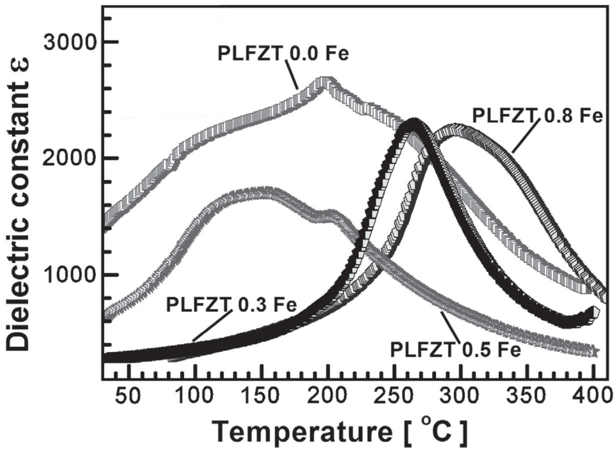
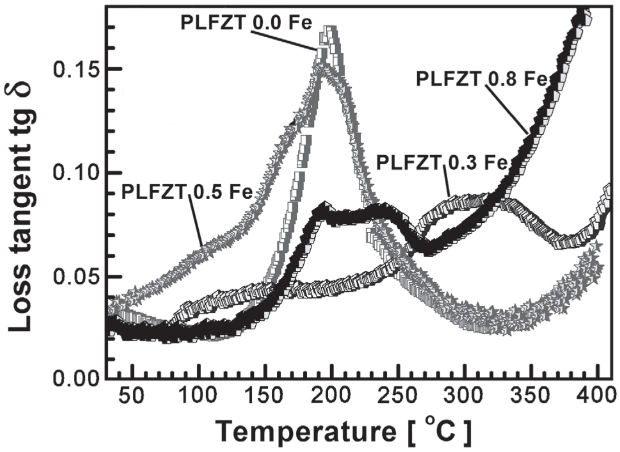


Figure 3. XRD diffraction patterns of $Pb_{0.91}(La_{1-x}Fe_x)_{0.09}(Zr_{0.65}Ti_{0.35})_{0.9775}O_3$ with: a) x = 0, b) x = 0.3, c) x = 0.5 and d) x = 0.8



a)



b)

Figure 4. Dielectric permittivity (a) and dielectric loss tangent (b) as a function of temperature at frequency of 1 kHz

ture. According to our and earlier published XRD investigation, rhombohedral *R3m* space group was assigned to the applied PLFZT composition (Fig. 3a,b,d) and coexistence of *R3m* and *P4mm* group for the PLFZT ceramics with $x = 0.5$ (Fig. 3c). From the measured and calculated profiles the crystallite size was derived using Pseudo-Voigt function. The lattice parameters were equal to a = 5.7406 Å and c = 14.1018 Å for the PLZT ceramics (x = 0) and gradually increased to the values of a = 5.7610 Å and c = 14.1670 Å for the PLFZT ceramics with x = 0.8. The final results refinement by Rietveld method for *R3m* phase for all samples revealed the PLFZT ceramics unit cell broadening, but the mentioned 1.2 % volume increase is not connected with symmetry lowering (c/a = const), implying that the Fe-addition in PLZT cause symmetric strain in the unit cell because of filling the interstices between octahedra in the A-site.

3.2 Electrical properties of PLFZT samples

The capacitance of the obtained PLFZT samples was measured at frequency of 1 kHz using a Quadtech 7600 LCR Meter for the calculation of dielectric coefficient (Fig. 4a). The dielectric loss coefficient was measured

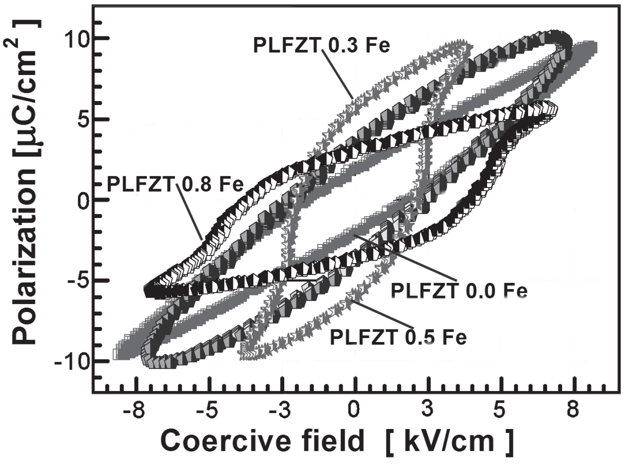


Figure 5. PLFZT ferroelectric hyseresis loops -polarization versus electric field (*P-E*) curves

at 1 kHz by the same method and the measured data are presented in Fig 4b. It is clearly seen, that the percentage of doping/substitution plays crucial role on the phase transitions and physical properties of the obtained materials. The temperatures of dielectric constant maxima were increased with increase of Fe^{3+} content in the PLFZT ceramics. A similar tendency in variation of loss tangent is also observed. As expected, a diffuse type of phase transition has been observed for x = 0 (Fig. 4a), but for higher x values the shape of dielectric constant curve become sharper in the maximum range indicating vanishing of relaxor behaviour. The level of dielectric loss values is very low and is not higher than 0.17 (for the PLFZT ceramics with x = 0.3) indicating good quality samples (Fig. 4b).

The ferroelectric hysteresis loops of all the investigated PLFZT compositions, from x = 0 to x = 0.8, are presented in the Fig. 5. As was supposed, each sample with lower Fe^{3+} content exhibits rounded shape loops with very low polarization of 2 μC/cm^2, whereas as Fe^{3+} intake increases, the hysteresis loops of these piezoelectric compositions become more quadratic in shape at room temperature, with relatively high polarization of 6 μC/cm^2.

3.3 Preparation and properties of piezoelectric transformers

All ceramic samples underwent the polarization process by applying the electric field of 3 kV/mm at the temperature of 150 °C for 15 minutes. After that, the piezoelectric transformers (PTs) were fabricated from the prepared PLZT and PLFZT ceramics. The bottom surface of PTs is covered by solid silver electrode, whereas opposite electrode patterns are deposited to create input and output parts separated by 2 mm insulating gaps, with electrodes' area ratio equal 1.66. The PTs were supplied with signal of 1 V in amplitude near the fundamental resonant frequency.

The AC magnetic field coils were driven by sinusoidal voltage generated by the function generator Hameg HM 8131 via the power amplifier HS2011. The sam-

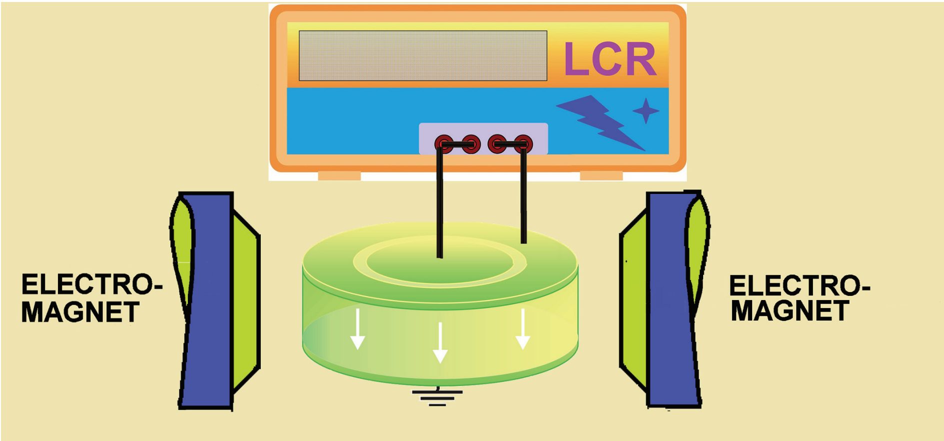


Figure 6. Scheme of impedance dependence on magnetic field intensity measurements setup

ple output impedance and phase values were measured by LCR Meter Quadtech 7600. All shown characteristics, namely ME voltage output as a function of the DC magnetic field, were obtained in the magnetic field of H_{AC} = 100 Oe at frequency of f = 1 kHz.

The measurement setup scheme is presented in Fig. 6. A computerized automatic system based on LABVIEW software was used to measure the PT electrical parameters dependency on the magnetic field intensity at frequency range near fundamental resonance.

Only the PLFZT composition with x = 0.5 exhibited non-zero magnetoelectric coupling and thus this particular ceramics was chosen for experiment with magnetic field sensing. This practically means that the coupling between magnetization and polarization is strong enough and this material become electrically polarized when placed in the magnetic field. The measured magnetoelectric coefficient was equal to 0.0115 mV/cm·Oe at f = 1 kHz and H_{AC} = 100 Oe. This value appeared to be sufficient to achieve magnetic influence on piezoelectric transformer performance. As was shown in Fig. 7a the frequency shift of 50 Hz and 100 Ω drop in PT output impedance under 100 Oe magnetic field was recorded. The impedance phase shift of 50 Hz is also detectable showing an alternative way to measure the intensity of the magnetic field (Fig. 7b).

The explanation of the high sensitivity of piezoelectric transformer for low ME coefficient lies in the fact, that piezoelectric transformer is a resonant operating device built from ferroelectric hard materials PLFZT with mechanical quality factor Q_m = 1000. Consequently, the output characteristic has very intense and narrow pattern with high sensitivity, and can thus be successfully used as a detector of magnetic field. Although, the previously reported application of piezoelectric transformer for magnetic field sensing [4] showed a 400 Hz frequency shift, the previously used material consisted of piezoelectric and ferromagnetic ceramics co-sintered together. In our particular case a single-phase multiferroic material was successfully implemented for the first time. Additionally, we demonstrated a simple and cost effective fabrication technique, as well as an easy sensing mechanism by detecting the frequency shift and/or the amplitude drop of the output signal due to the impedance change.

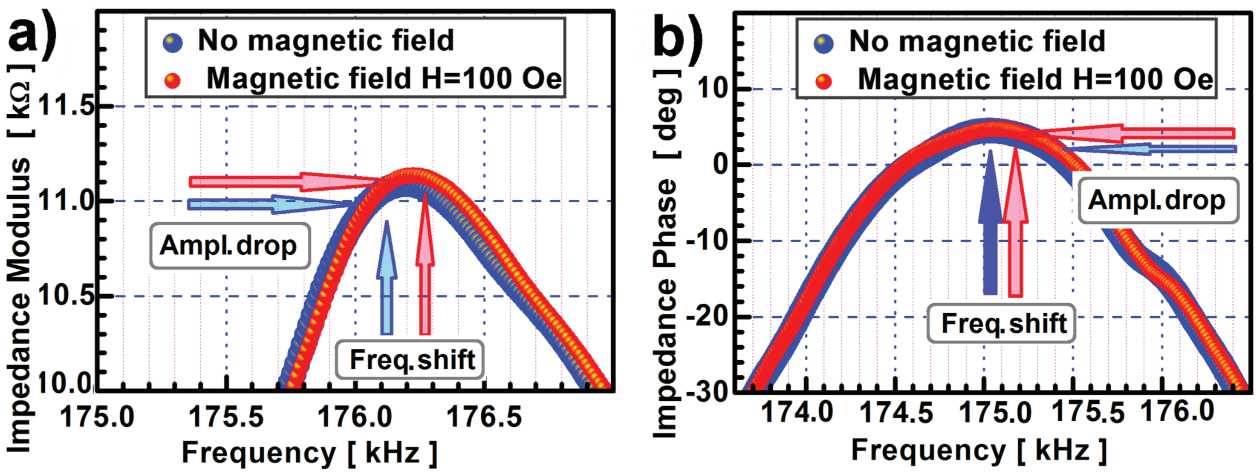


Figure 7. Piezoelectric transformer with magnetic feedback characteristics. The magnetic field induced frequency shift in impedance modulus (a) and phase maxima (b) together with respective amplitudes drop.

IV. Conclusions

The present report introduces a new method of magnetic field-sensing detection using the piezoelectric transformer prepared from multiferroic material. Among investigated compositions, the $Pb_{0.91}(La_{0.5}Fe_{0.5})_{0.09}(Zr_{0,65}Ti_{0,35})_{0,9775}O_3$, located near to the morphotropic boundary, exhibits the highest magnetoelectric effect, probably due the high values of piezoelectric coefficients increasing the electrical response for magnetic field induced strain. Good quality PLFZT material, with ferroelectric and ferromagnetic properties at the same time, can be prepared by pressure assisted high temperature sintering technique with Fe^{3+} substitution at the lanthanium sites.

From the dielectric constant temperature dependencies it is clearly seen, that percentage of Fe^{3+} doping/substitution plays a crucial role in the phase transitions and physical properties of the obtained materials.

Finally, the $Pb_{0.91}(La_{0.5}Fe_{0.5})_{0.09}(Zr_{0,65}Ti_{0,35})_{0,9775}O_3$ ceramics was implemented into piezoelectric transformer construction and demonstrated the effectiveness of the discussed magnetic field detecting technique. The development and study of this application might be the fundamental basis for the practical "cheap detector" design, in which the output parameters value might be measured using conventional multimeters instead of high level lock-in technique.

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