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Multilayered ceramic heterostructures of lead zirconate titanate and nickel-zinc ferrite for magnetoelectric sensor elements

D.Yu Karpenkov[a], A.A. Bogomolov[b], A.V. Solnyshkin[b,c], A.Yu Karpenkov[b], V.I. Shevyakov[c], A.N. Belov[c,*]

[a] The National University of Science and Technology “MISiS”, 119991 Moscow, Russia
[b] Tver State University, 170100 Tver, Russia
[c] National Research University of Electronic Technology, Zelenograd, 124498, Moscow, Russia

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ABSTRACT

The preparation method of magnetoelectric multilayered ceramic heterostructures was suggested, and series of the rectangular and disk shape samples were produced on the base of this method. The maximum value of a magnetoelectric voltage coefficient $\alpha_E = 35$ V/(cm × Oe) was observed for the eleven-layered rectangular specimen at the electromechanical resonance frequency $f_r = 144$ kHz, $H_{DC} = 84$ Oe, and $H_{AC} = 2.5$ Oe. The sensitivity to static magnetic fields is equal to 3.5×10^{-2} Oe, and the alternating field sensitivity is equal to 8×10^{-3} Oe.

1. Introduction

In the last decade, much attention was paid to the magnetoelectric (ME) effect in multilayered composite structures containing layers of magnetostrictive Ni-Zn ferrite (NZFO) and piezoelectric (PZT) materials [1–11]. This interest is caused by the great value of the ME coefficient in multilayered structures and potential applications in magnetic field sensors [12] and solid-state voltage converters [13], as well as the relatively easy manufacturing process of multilayered composites. However, reproducibility of ME parameters is not adequate, that prevents widespread application of the devices based on the layered heterostructures. Furthermore, achieving high ME coefficient values requires a good mechanical coupling, as well as chemically nonreactive phases of the composite.

In this paper, we propose a new preparatory method for ME structures consisting from NZFO and PZT layers. This method eliminates the voltage drop across the dielectric layers of magnetostrictive ceramics, and also allows the combination of layers of PZT ceramics electrically in series, which subsequently increases the efficiency of the composites. The results of systematic investigations of the ME effect for the samples produced by the proposed method are presented. The ME voltage coefficient is determined in wide ranges of applied magnetic fields, frequencies, and temperatures.

2. Magnetoelectric heterostructures

We investigated two-, three-, five-, and eleven-layered samples of the ceramic heterostructures. As a piezoactive material, the commercial ceramic plate PZT-46 (Elpa Company, Russia) was used. Plates of nickel-zinc $Ni_{0.8}Zn_{0.2}Fe_2O_4$ ferrite were used as a magnetostrictive layer. The choice of ferrite with such composition is determined by the magnetostriction λ, the piezomagnetic coefficient $q = d\lambda/dH$, and permeability μ defining the strength of direct and converse magnetoelectric coupling. Heterostructures consisted of (n=1) layers of piezoceramic (rectangular samples of size 18×9×0.13 mm and disk-shaped samples of 18 mm diameter and 0.1 mm thickness) and n layers of ferrite (with the same linear sizes as above and 0.2 mm in thickness) (n = 1, 2, 5). Two types of geometries were chosen to investigate the influence of the shape of whole structure on its performance. It is well known that in disk-shaped actuators the radial modes are observed instead of rectangular one, which reveals longitudinal and transverse modes of vibrations. Moreover for practical application it is necessary to found out the impact of compress effect from magnetostrictive layers on the magnetoelectic response in both structure geometries.

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* Corresponding author at: National Research University of Electronic Technology, Zelenograd, 124498, Moscow, Russia.
E-mail address: nanointech@mail.ru (A.N. Belov).

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The created ceramic heterostructures can be considered as the batteries of PZT capacitors connected in series. Each PZT element in the battery is mechanically rigidly coupled with the ceramic NZFO magnetoactive layers. The entire heterostructure is mechanically monolithic. For such a composite ceramic structure the following can be measured: 1 – the potential difference between the upper and lower electrodes of the heterostructure; and 2 – resonance and antiresonance frequencies generally determined by vibration modes that are explicit functions of dimensions and elastic characteristics of the entire heterostructure.

In the presence of an alternating magnetic field, the sizes of ferrite change due to magnetostriction, which induces: 1 – the mechanical strain of the PZT element that, in turn, leads to the appearance of the voltage across each PZT element and the total potential difference between the upper and lower electrodes of the whole heterostructure; and 2 –changes in the elastic characteristics of the heterostructure and its dimensions, which causes the shift of resonance and antiresonance frequencies of different vibration modes.

The number of layers was chosen to obtain the maximum value of the magnetoelectric response of the heterostructure and optimal technical parameters required for the magnetic fields sensors. The conductive epoxy glue CW2460 was used to achieve strong mechanical coupling between the layers.

To improve electrical contact between the PZT layers, the entire silver electrodes were deposited on the whole surface of ferrite plates. In this case, the ferrite plates were switched off from an electrical circuit of the heterostructures as a dielectric layer.

Despite many attempt to produce practically efficient multilayred heterostructured based on Ni-Zn ferrite, the majority of them are bi- or three-layered [18–24]. It is mostly caused by dielectric properties of the magnetoactive layer. However, to increase the ME response of the laminates it is necessary to connect all piezoelectric layers in series. The proposed method of heterostructure compacting eliminates the voltage drop across the dielectric layers of magnetostrictive ceramics due to the presence of a conductive coating on the side surface that forms an electric contact of the PZT plates. This increases the efficiency of magnetic-to-electric energy conversion for the multilayered heterostructures created as the basis for the magnetoelectric sensor elements. Extended description of the developed method of multilayered ceramic heterostructure production can be found in [25].

3. Experimental technique

To investigate the ME effect, a method based on measuring the AC voltage generated by the composite sample under magnetic fields was used. In the presence of a static magnetic field corresponding to the maximum ME effect, the frequency dependence of the magnetoelectric response was measured in the range from 17 Hz to 230 kHz. The DC magnetic field dependence on the ME coefficient in the range of 0–1500 Oe was investigated at a fixed amplitude of the alternating magnetic field $H_{AC} = 2.5$ Oe and at the resonance frequency of the main longitudinal vibration mode. The measurements were performed for the longitudinal orientation of static and alternating magnetic fields. H_{DC} and H_{AC} vectors were collinear and oriented in the plane of the heterostructure. The magnetoelectric voltage coefficient was calculated on the basis of following equation:

$$\alpha_E = \frac{E_3}{H_1} \tag{1}$$

where H_1 is the amplitude of the AC magnetic field and E_3, the electric field in the material, is determined according to:

$$E_3 = \frac{U_0}{d} \tag{2}$$

where d is the thickness of the PZT material, $U_0 = U/k$, U is the output voltage of the amplifier, and k is the gain coefficient.

Magnetostriction measurements were carried out with the use of SK-06-030TY-350 strain gauge sensors (Vishay Company, Wendell, USA). The experimental technique and apparatus for the measurement of magnetostriction was described in [14]. During the measurements, the samples were mechanically free. This state was provided by means of point contacts to opposite surfaces of the multilayered structures.

4. The results and discussion

Fig. 1(a) and (b) show the frequency dependence of the AC voltage generated by the heterostructure samples of rectangular shape and disk shape, respectively. The measurements were performed for rectangular specimens under the static magnetic field $H_{DC} = 84$ Oe, and for the disk-shaped samples under $H_{DC} = 200$ Oe, which corresponds to the maximum efficiency of the ME interaction.

The amplitude of the alternating magnetic field applied was constant over the entire frequency range and equal to $H_{AC} = 2.5$ Oe for rectangular shape samples and $H_{AC} = 0.3$ Oe for disk-shaped samples. While the frequency of the external alternating magnetic field was changing, the resonances of magnetoelectric response occurred. This is caused by an excitation of proper acoustic vibration modes of the structure, which leads to a high resonant amplitude of deformations and, consequently, increases the ME response [15]. According to our data, the maximum value of the ME response was observed at a second harmonic of a planar vibra-

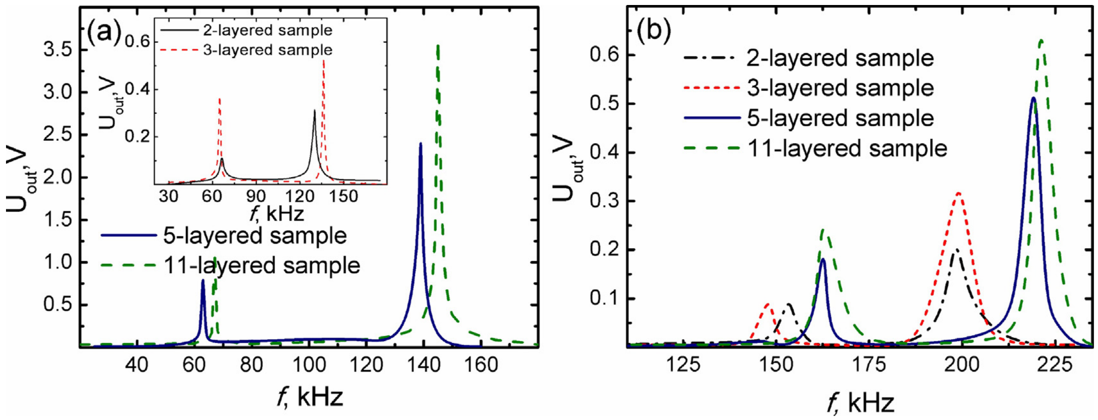

Fig. 1. The frequency dependence of ME voltage for: (a) rectangular-shaped samples, or (b) disk-shaped samples.