

Dependence of magnetoelectric effect in layered lead zirconate-titanate / nickel heterostructures on the interface type

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Results for the dependence of low-frequency magnetoelectric (ME) voltage coefficient on the Ni/PZT structure type and contact preparation method are presented. Different type of structure are applied by ion-beam evaporation, electrolytic deposition, and epoxy adhesive connection of nickel plate and lead zirconate titanate (PZT) polarized ceramics. The shift of the maximum ME effect towards weaker magnetic field observed in experiment is accompanied by the increase of the coercive force and a change in mechanical Q-factor of composites. The comparison of ME coefficient for the bilayer and three-layer structures has shown a increase in the ME response for Ni/PZT samples.

Представлены результаты исследования зависимости низкочастотного магнитоэлектрического МЭ коэффициента по напряжению от типа структуры и метода соединения структур Ni/ЦТС. Формирование металлических структур проводилось ионно-лучевым распылением, электрохимическим осаждением и механическим соединением пластин никеля с поляризованной керамикой ЦТС. Экспериментально наблюдаемое смещение максимума МЭ коэффициента в область меньших магнитных полей сопровождается ростом коэрцитивной силы образцов и изменением добротности Q. Сравнение МЭ коэффициента двух- и трехслойных структур показало возрастание МЭ отклика для образцов Ni/ЦТС.

1. Introduction

The direct magnetoelectric effect (ME) consists in the appearance of an electric field E in structures coupled by exchange interaction which have been placed into an external magnetic field H [1]. In composite materials, polarization of a piezoelectric component appears due to the mechanical transfer of striction from a magnetostrictive phase which is subjected to the external magnetic field [2]. This results in the appearance of a voltage U at the structure surfaces.

The value of the ME response of a composite structure is defined both by the features of the magnetic field acting upon the structure (values, orientations and frequencies of the alternating field) and by the composite structure properties. The interest of researchers in layered magnetoelectric materials which has intensified in recent years [3–5] is due to numerous reasons. The material polarization is not restrained by the magnetostrictive component conductivity. In such structures, it is possible to use pure metals with magnetostrictive properties several orders of magnitude higher than magnetostriction of ferrites. A possibility is appeared to combine various methods to form the magnetic and piezoelectric phases. In this case, the magnetic phase structure and the properties of the contact at the heterostructure boundaries will effect on the ME response of the obtained composite material.

2. Preparation of samples and research methods

This work presents the experimental study results on the dependence of low-frequency ME voltage coefficient in metal-piezoceramics structures in the external magnetic field up to 2 kOe on the metallic phase type (thin film, thick layer, nickel plate) and the kind of its contact with ferroelectric ceramics. The lead zirconate-titanate (PZT 42) disks of 8.5 mm in diameter thinned to 500 μm thickness with both surfaces polished to the rms roughness of less than 100 nm were used as piezoelectric substrates. Thin nickel films of up to 2.5 μm thickness were deposited by the ion-beam evaporation. The nickel layers were obtained by forming a seed contact layer of up to 0.5 μm thickness by ion-beam evaporation followed by electrolytic deposition of nickel films up to the preset layer thickness. The mechanical connection of nickel foil disks with a thickness up to 500 μm and PZT was provided by epoxy resin application followed by pressing and drying, resulting in a contact of ferromagnetic/piezoelectric bulk phases. The approach described provided observation of the ME response peak value in each case due to ensuring maximum magnetostrictive properties of the obtained ferromagnetic phase structure [6, 7]. The study was carried out in dynamic conditions. The samples were placed in alternating and constant magnetic fields and the voltage change appearing in the structure was measured. The ME voltage coefficient dE/dH was determined based on the sample thickness h, the voltage rate dU and the alternating magnetic field strength dH: $dE/dH = dU/(h{\cdot}dH)$. The research at 1 kHz frequency was carried out for transverse effect when the sample electric polarization vector is directed transversely to external magnetic fields.

The selection of the layered heterostructure materials is conditioned by the intrinsic properties of ferromagnetic and piezoelectric components. The main advantage of nickel is the saturation of magnetostriction in the external field of less than 0.5 kOe. Besides, nickel is convenient in processing, has a low oxidation rate and is suitable to formation of various type coatings. The PZT powder ensures good piezoelectric properties of workpieces during sintering and processing by traditional and available ceramic technologies. After the ceramic synthesis, the PZT workpieces were polarized in silicone fluid for two hours at 150°C in the field 4 kV/mm with the subsequent cooling in this field.

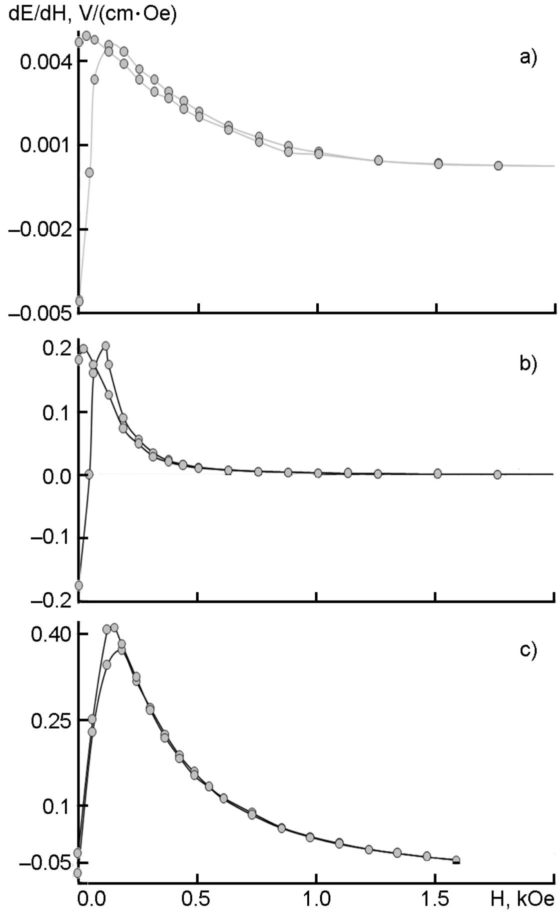

Fig. 1. Field dependences of the low-frequency ME voltage factor in three-layered structures obtained by ion beam evaporation (a), electrolytic deposition (b), and mechanical coupling of the components.

3. The study results and discussion

The field dependences of three-layer Ni/PZT/Ni structures are shown in Fig. 1. The plot *a* in Fig. 1 describes the behavior of the low-frequency ME voltage coefficient for the structure obtained by ion-beam evaporation of nickel film. This method of metallization provides contact between phases of a composite at the nanoscale level [8]. The nickel layer thickness at each side was 2 μm.

The dependence of the ME voltage coefficient for a sample with 30 μm thick nickel

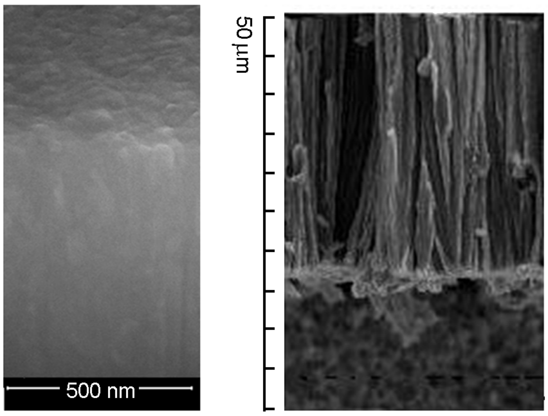

Fig. 2. The cross-section microstructure of Ni/PZT samples with nickel layer obtained by ion beam evaporation (left) and electrochemical technique (right).

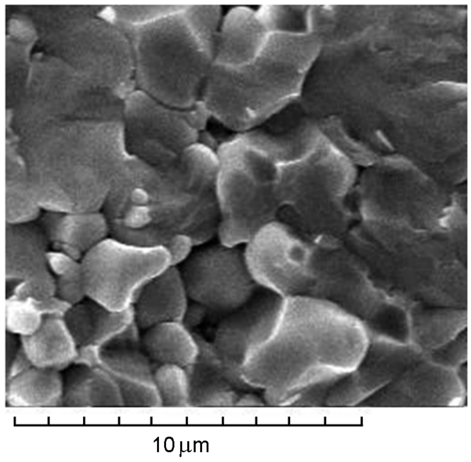

Fig. 3. Microstructure of piezoelectric PZT 42 ceramics.

layers at each side prepared by electrolysis is presented by the plot *b* in Fig. 1. The electrolyte composition and the deposition modes were selected according to the data [6] to obtain the maximum magnetostriction at the largest possible film thickness. According to the models discussed in [6], the nickel magnetostriction increment with the increase of ferromagnetic layer thickness is provided due to formation of the columnar layer structure. The view of the investigated heterostructure cross-section is shown in Fig. 2.

Fig. 1, c describes the ME voltage coefficient behavior for a structure where the ceramic substrate is in mechanical contact with nickel at both sides. The magnetostrictive disks of cast nickel were annealed for two hours in hydrogen medium at a temperature of 500°C. The structure of the bulk ferromagnetic phase in this case is isotropic and has minimal internal stresses as compared to other investigated types.

The experimental research has shown that structures with thin nickel layers exhibit the greatest coercive force (44 Oe) (Fig. 1, a). The coercive force decreases to 39 Oe in the structures with nickel layers (Fig. 1, b). The internal stresses in the electrolytic films is much lower than those in thin nickel films obtained by the ion-beam evaporation, thus allowing the magnetostrictive coating thickness to be increased up to tens of micrometers. In the samples connected mechanically (Fig. 1, c), the annealed nickel disks possess minimum internal stresses, as a result, the disks have minimal coercive force up to 5 Oe.

The observed shift of the ME coefficient maximum towards weaker magnetic fields accompanied by the increase in the sample coercive force is explained by the dependence of nickel layer properties on its formation method by and the peculiarities of the boundaries between the ferromagnetic and piezoelectric phases in each investigated case. The microstructure images of the nickel layer obtained by electrolysis show that the average width of its transverse structure columns is 150 nm. In the ion-beam evaporated films, the maximal width of the columns does not exceed 110 nm. The described coating structure changes are in correlation with the grain size of PZT 42 ceramics (Fig. 3) and define the phase contact total area. This results in the observed peculiarities of the change in magnetoelectric properties. This conclusion is further confirmed basing on the ME characteristics comparison for the heterostructures of three-layer (described above) and bilayer (Ni/PZT) type.

The ME response in the three-layer samples obtained by the electrolytic deposition of 55 µm thick nickel films at each side was 140 mV/(cm·Oe). This value exceeds by a factor of 1.5 the ME effect value for the bilayer samples, the ferromagnetic to piezoceramics volume ratio being the same. In case of the mechanical contact, the ME effect value for the three-layer structures corresponded to 390 mV/(cm·Oe). In the bilayer samples, it was 190 mV/(cm·Oe). The thickness of each nickel disk was 125 µm for the three-layer sample and 250 µm for the bilayer one.

In experiment, a greater ME voltage coefficient is observed in the three-layer samples for all the studied methods of layered materials preparation as compared to the bilayer structures. This means that in the ferromagnetic-piezoelectric-ferromagnetic heterostructures, the response to the same change of the external magnetic field expressed as the electric field increment exceeds at least by 1.5 times the response observed in the piezoelectric-ferromagnetic heterostructures under the experimental conditions described above. The induced electric field value in a sample depends only on the strain of piezoceramics defined by the change of ferromagnetic metal dimensions. The ferromagnetic metal straining, in its turn, is defined uniquely by the value of the applied magnetic field. For this reason, one can suppose that redistribution of the sample relative strain occurs both in bilayer and three-layer structures. Most probably The ME properties of Ni/PZT/Ni type structures are defined most likely by longitudinal strains of the sample. For bilayer Ni/PZT heterostructures, radial and bending strains are essential.

The mechanical Q factor was determined at the frequency of ME resonance. In ion-beam evaporated thin nickel films, its magnitude attained 1200. At the mechanical connection of nickel disks and piezoceramics, it corresponded to 150. In the samples obtained by electrolytic deposition, Q increased from 300 to 700 when nickel layer thickness decreased from 60 μm to 30 μm.

It is known that the stronger is the interconnection between the components of the ferromagnetic/piezoelectric structures, the higher is the Q-factor of composite structures [5]. The strongest interconnection between the heterostructure phases is ensured in case of ion-beam evaporation of thin nickel films onto PZT ceramics. As a result, the Q-factor attains values comparable with the Q-factor of bulk composite materials. At the mechanical contact of phases, there is no direct contact between the piezomaterial and ferromagnetic, so the Q-factor is minimum in such structures.

4. Conclusions

The experimental study of the PZT/Ni heterostructures has shown a correlation between coercive force value, ME response shift, Q-factor, and the type of layered structure. In all the cases, the observed peculiarities in magnetoelectric properties changes is explained by the dependence of the magnetostrictive nickel layer properties on its formation method and on the coupling at the ferromagnetic/piezoelectric phase interface. The described coating structure changes are in correlation with piezoceramics grain size and define the phase contact total area. The measured Q-factor values for the studied materials also show an essential influence of near-boundary interaction on the ME response behavior of the composite material. Such a conclusion is confirmed when comparing the ME characteristics of the Ni/PZT/Ni and Ni/PZT heterostructures. In this case, the interconnection of magnetostrictive and piezoelectric components in the near-boundary layer acquires a decisive role resulting in significant losses in the longitudinal ME voltage coefficient value.

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Залежність магнітоелектричного ефекту у шаруватих гетероструктурах цирконат-титанат свинцю/нікель від типу поверхні контакту

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Подано результати дослідження залежності низькочастотного магнітоелектричного (МЕ) коефіцієнта за напругою від типу структури та способу з'єднання структур Ni/ЦТС. Формування металевих структур виконувалося іонно-променевим розпиленням, електрохімічним осадженням та механічним з'єднанням пластин нікелю з поляризованою керамікою ЦТС. Зсув максимума МЕ коефіцієнта в область менших магнітних полів, що експерементально спостерігався, супроводжується зростанням коерцитивної сили зразків та зміною добротності Q. Зіставлення МЕ коефіцієнтів для дво- та тришарових структур показало зростання МЕ відгуку для зразків Ni/ЦТС.