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THE TECHNOLOGY OF MANUFACTURE AND MAGNETOELECTRIC CHARACTERISTICS OF STRUCTURE, OBTAINED BY GALVANIC DEPOSITION OF TIN AND NICKEL ON A SUBSTRATE OF GALLIUM ARSENIDE

D.A. Filippov[1*], **I.N. Manicheva**[1]

[1]Department of Engineering Technology, Novgorod State University, Velikiy Novgorod, Russia

Abstract. The technology of fabrication and the results of the magnetoelectric effect investigation in sandwich structure manufactured by galvanic deposition tin and nickel on the gallium arsenide substrate are presented. It is shown that the use of tin as an intermediate layer lead to reduces the mechanical stresses resulting on the interface nickel and gallium arsenide. It is possible to obtain qualitative structures with nickel layer thickness on the order of 100 microns. Experimental results of the frequency dependence of the magnetoelectric voltage coefficient in the region of electromechanical resonance are presented. The resonance value of the magnetoelectric voltage coefficient reached 40V/ (cm Oe) with the Q-factor ≅ 700, which significantly exceeds the characteristics of similar structures obtained by bonding.

Keywords: *magnetoelectric effect, gallium arsenide, nickel, tin, adhesion, galvanic deposition*.

Corresponding Author*: Dmitry Filippov, Professor, Department of Engineering Technology, Novgorod State University, 173003, ul. B.St.Peterburgskaya, 41, Velikiy Novgorod, Russia, Tel.: (81622) 626429, e-mail: Dmitry.Filippov@novsu.ru*

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1. Introduction

Composite magnetostrictive-piezoelectric materials attract attention due to the fact that the magnetoelectric (ME) effect is possible in them, which consists in changing the polarization of the sample when placed in a magnetic field. The magnitude of the ME effect in composite materials is several orders of magnitude greater than in single crystals. This is explained by the fact that the mechanism of the ME effect in single crystals is the joint action of the spin-orbit interaction, the interaction of an electron with an external and crystalline electric field (Bichurin & Filippov, 1997). Despite the fact that dozens of single crystals are now known in which the ME effect (Zvezdin & Pyatakov, 2012) is detected, nevertheless, owing to its smallness, single crystals have not found wide application in engineering. Mechanism of occurrence of ME effect in composites is mechanical interaction of magnetostrictive and piezoelectric phases. The alternating magnetic field causes mechanical deformations in the magnetostrictive component, which are transferred through the interface to the piezoelectric component, which leads to a change of polarization and the appearance of an electrical voltage. Composite ME materials can be divided into groups: bulk and layered composites. Bulk composites are mechanically bound mixtures of powders of magnetic and piezoelectric phases (Laletin & Srinivasan, 2002). Layered composites are structures consisting of alternating layers of a magnetic and a piezoelectric (Srinivasan *et al*., 2001). Layered structures have several advantages over bulk composites: they are easily polarized, they have small leakage currents (Filippov *et al.*, 2012), metals with a large magnetostrictive coefficient can be used as the magnetostrictive phase (Fetisov, 2009, 2007; Filippov, 2016). At the same time, they have a number of disadvantages due to the interface between the magnetostrictive and the piezoelectric phases. Most layered ME structures are obtained by bonding, which leads to a decrease in the quality factor of the structure, weakening of the ME effect, undesirably high temperature dependence. In (Gridnev *et al.*, 2015; Laletin, 2014) the results of an investigation of the ME effect in structures were reported, where the magnetostrictive phase was applied to the piezoelectric substrate by the sputtering method. The resulting structures had good adhesion, but had a small value of the ME effect. As shown in (Filippov *et al.*, 2013) the maximum of the ME effect is observed under the condition of equality ${}^{p}t\sqrt{{}^{p}Y} = {}^{m}t\sqrt{{}^{m}Y}$, where ${}^{p}Y$, ${}^{m}Y$ are the Young's moduli of the piezoelectric and magnetic phases, and ${}^{p}t$, ${}^{m}t$ are their thickness. Since the Young's moduli of a magnetic and a piezoelectric phase do not differ by more than a factor of two, the maximum effect is observed when the thickness of the magnetic phase is commensurable with the thickness of the piezoelectric layer. It is impossible to fabricate such structures by the sputtering method. Using the method of electrolytic deposition of metal on the piezoelectric substrate allows one to obtain layers of a magnetic phase whose thickness is commensurable with the thickness of the piezoelectric phase. In order to improve the adhesion between phases during fabrication, it is advisable to use structures preliminarily deposited on a GaAs substrate by Au-Ge-Ni sublayers (Filippov *et al.*, 2017). However, due to the incommensurability of the lattice parameters of Ni and GaAs at large layer thicknesses, mechanical stresses arise, leading to warping of the structure and its destruction. One of the methods for eliminating these stresses, proposed in this paper, is the method of creating a sandwich structure in which the nickel layer alternates with a buffer layer of tin.

2. Technology of manufacturing structures

Samples in the form of a parallelepiped with dimensions $11 \times 5 \times 0.4$ mm were cut from GaAs plates with a surface orientation (100), the long side of which coincided with the direction of the <011> crystal (Figure 1). To improve the adhesion to the samples, Au-Ge-Ni sublayers were previously deposited. Before plating, all samples were first contacted with a 0.2 mm diameter nickel wire. The electrolytes shown in Table1 were used for the application of galvanic coatings.

Table 1. The composition of electrolytes used to create structures

Components of electrolyte, g/l	Electrolyte №1	Electrolyte №2
Nickel sulfate heptahydrate	250	-
Nickel chloride hexahydrate	50	-
Tin sulfate	-	60
Boric acid	25	-
Sulfuric acid	-	105
The preparation OS-20	-	4.5

In the production of the multilayer structure, electrolytic deposition was used alternately in the sulfuric acid electrolyte of nickel №1, with a cathode current density of 1 A/dm^2 and an electrolyte temperature of 55-65 °C, and then electrodeposition in the tin electrolyte №2 at room temperature and a cathode current density of 2 A/dm^2. As a result, the resulting multilayer structure consisted of sixteen layers of nickel on each side of the sample, with a total thickness of 100 μm and seventeen layers of tin, with a total thickness of 200 μm. The total thickness of the sandwich structure, taking into account the substrate, was 1 mm. The layers on gallium arsenide had an even, matte surface, without visible defects.

3. Magnetoelectric effect

The magnetoelectric (ME) effect in the structure was studied by measuring the voltage on the sample when placed in a constant (magnetizing) and alternating magnetic fields directed along the long side of the sample (Fig. 1).

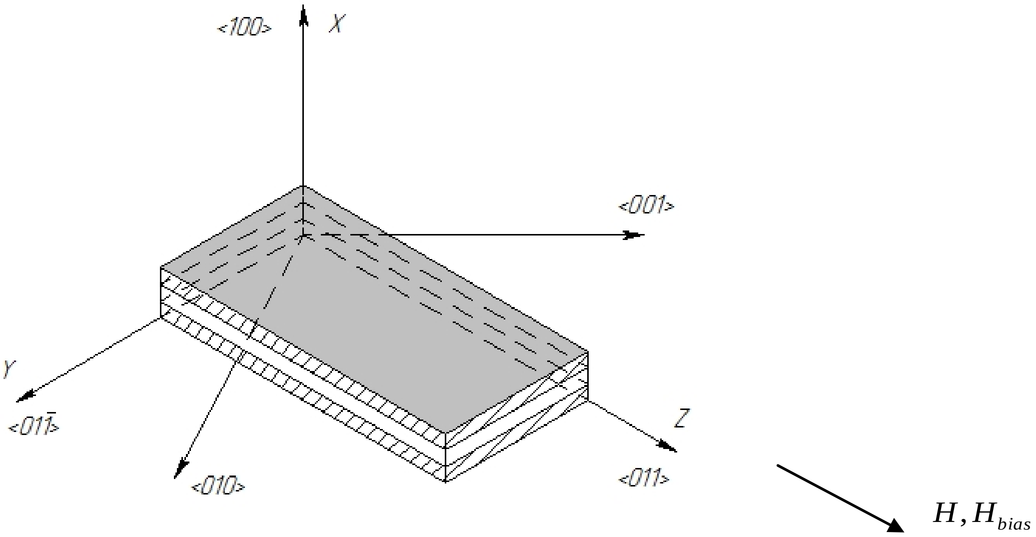

Figure1. The schematic drawing of the sandwich structure

First, the field dependence of the low-frequency ME signal was investigated. At a constant value of the strength of the alternating magnetic field $H=1\ Oe$, the dependence of the ME coefficient on the strength of the bias field H_{bias} was measured. Then, with the magnetization field strength corresponding to the maximum of the effect, the frequency dependence of the ME coefficient in the electromechanical resonance region was investigated. In gallium arsenide have nonzero components $d_{14}= d_{25}= d_{36}$ of the piezoelectric tensor, therefore, the electric voltage induced on the sample plates arises from shear deformations rather than deformations tension-compression as in PZT (Filippov *et al.*, 2013). The alternating magnetic field directed along the long side of the sample (the Z axis) induces deformations of tension-compression in the magnetic component whose tensor, in the coordinate system associated with the sample, will be denoted by S_{zz}. These deformations are transferred to the piezoelectric, which leads to the appearance of an electrical voltage on the plates. To find the ME characteristics of the sandwich structure, we use the method developed earlier for a two-layer structure (Filippov *et al.*, 2014). We use the fact that the thickness of the piezoelectric, magnetic and buffer layer is much smaller than the length of the sample, so in the first approximation it can be assumed that the displacements of the layers are the same and do not vary in the thickness of the sample. In this approximation, the equation of motion for the z-projection of the displacement vector of the medium is written in the form:

$$\bar{\rho}\frac{\partial^2 u}{\partial t^2} = \frac{\partial \bar{T}_{zz}}{\partial z}, \tag{1}$$

where $\bar{\rho}$ is the mean value of the sample density, and $\bar{T}_{zz}$ is the mean value of the stress tensor in the sample.

The equations for components of the strain tensor of piezoelectric ${}^{p}S_{zz}$ and magnetic ${}^{m}S_{zz}$, and electric induction ${}^{p}D_{x}$ are as follows:

$${}^{p}S_{zz} = \frac{1}{{}^{p}Y}\,{}^{p}T_{zz} + {}^{p}d_{x,zz}\,{}^{p}E_{x}, \tag{2}$$

$${}^{m}S_{zz} = \frac{1}{{}^{m}Y}\,{}^{m}T_{zz} + {}^{m}q_{z,zz}\,{}^{m}H_{x}, \tag{3}$$

$${}^{p}D_{x} = {}^{p}\varepsilon_{xx}\,{}^{p}E_{x} + {}^{p}d_{x,zz}\,{}^{p}T_{zz}, \tag{4}$$

where ${}^{p}T_{zz}$, ${}^{m}T_{zz}$ are components of the stress tensor in the piezoelectric and magnetostrictive phase; ${}^{p}Y$, ${}^{m}Y$ are the Young`s modulus in the piezoelectric phase along the <011> (Z-axis) direction, and the magnetic phase, respectively, ${}^{p}d_{x,zz}$ is the piezoelectric tensor in the XYZ coordinate system (Figure 1), ${}^{p}\varepsilon_{xx}$ is the dielectric tensor and ${}^{p}E_{x}$ is the x component of the electric field strength vector, ${}^{m}q_{z,zz}$ is the piezomagnetic coefficient, ${}^{m}H_{x}$ is the magnetic field strength.

The components of the piezoelectric tensor ${}^{p}d_{x,zz}$ in the *XYZ* coordinate system are related to the components of the piezoelectric tensor ${}^{p}d_{\alpha,\beta}$ in the crystallographic coordinate system by the relation:

$$d_{x,zz} = d_{14}\beta_{z2}\beta_{z3}, \tag{5}$$

where β_{z2}, β_{z3} is the matrix of cosines between the Z axis and the axes 2 and 3 (<010> and the direction <001>).

The solution of equation (1) for the displacement vector of the medium is represented in the form of plane waves propagating along the length of the sample:

$$u(z) = A\cos(kz) + B\sin(kz), \tag{6}$$

where A and B are the integration constants.

Substituting expression (6) into equation (1), we obtain for them the dispersion relation in the form:

$$\omega = \sqrt{\frac{\bar{Y}}{\bar{\rho}}}\,k = \sqrt{\frac{{}^{m}Y\,{}^{m}t + {}^{l}Y\,{}^{l}t + {}^{p}Y\,{}^{p}t}{{}^{m}\rho\,{}^{m}t + {}^{l}\rho\,{}^{l}t + {}^{p}\rho\,{}^{p}t}}\,k, \tag{7}$$

where $\bar{Y} = \dfrac{{}^{m}Y\,{}^{m}t + {}^{l}Y\,{}^{l}t + {}^{p}Y\,{}^{p}t}{{}^{m}t + {}^{l}t + {}^{p}t}$ the average Young's modulus of the sandwich structure.

4. Magnetoelectric voltage coefficient

The magnetoelectric voltage coefficient is defined as the ratio of the average intensity of the electric field <E> to the intensity of the alternating magnetic field H

$$\alpha_E = \langle E \rangle / H \,, \tag{8}$$

where $\langle E \rangle = U / ({}^{m}t + {}^{l}t + {}^{p}t)$ is the average value of the electric field in the structure and U is the voltage induced between the electrodes.

To obtain the expression for the ME coefficient, we use the method developed earlier in (Filippov, 2013, 2014). From the condition of mechanical equilibrium at the free sidewalls of the sample, i.e. at the points $z = \pm L/2$ we have the following boundary conditions:

$$\int_0^{{}^{p}t} {}^{p}T_{zz}(\pm L/2, \mathrm{x})\,dx + \int_{{}^{p}t}^{{}^{l}t} {}^{l}\mathrm{T}_{zz}(\pm L/2, x)dx + \int_{{}^{l}t}^{{}^{m}t} {}^{m}T_{zz}(\pm L/2, x)dx = 0 \tag{9}$$

Using these boundary conditions, for integration constants we obtain:

$$A = 0,$$

$$B = \frac{{}^{m}Y\,{}^{m}t\,{}^{m}q_{\mathrm{z,zz}}\,{}^{m}H_x + {}^{p}Y\,{}^{p}t\,{}^{p}d_{\mathrm{x,zz}}\,{}^{p}E_x}{k\cos(\kappa)} \frac{1}{({}^{m}t\,{}^{m}Y + {}^{l}t\,{}^{l}Y + {}^{p}t\,{}^{p}Y)}, \tag{10}$$

where a dimensionless parameter $\kappa = kL/2$ is introduced. Having expressed the component of the stress tensor in terms of the components of the strain tensor from Eq. (2) and substituting the resulting expression into the equation for the normal component of the electric induction vector, we obtain to the expression in the form:

$${}^{p}D_z = {}^{p}\varepsilon_{xx}\,{}^{p}E_x + {}^{p}Y\,{}^{p}d_{x,zz} \frac{\partial\,{}^{p}u_z}{\partial z} - {}^{p}Y({}^{p}d_{x,zz})^2\,{}^{p}E_x \,. \tag{11}$$

To determine the electric field ${}^{p}E_x$ induced in a piezoelectric, we use of the open-circuit condition:

$$I = \iint \frac{\partial D_x}{\partial t} dzdy = 0 \,. \tag{12}$$

Substituting expression (11) into equation (12) and carrying out the integration, we obtain:

$${}^{p}E_x = \frac{{}^{p}Y\,{}^{p}d_{\mathrm{x,zz}}\,{}^{m}q_{\mathrm{x,zz}}}{{}^{p}\varepsilon_{xx}} \frac{{}^{m}H_x}{\Delta} \frac{{}^{m}Y\,{}^{m}t}{({}^{\mathrm{m}}\mathrm{t}\,{}^{\mathrm{m}}Y + {}^{l}\mathrm{t}\,{}^{l}Y + {}^{p}\mathrm{t}\,{}^{\mathrm{p}}Y)} \frac{\tan(\kappa)}{\kappa} \,, \tag{14}$$

where the notation

$$\Delta = 1 - k_p^2 \frac{{}^{p}t\,{}^{p}Y}{({}^{m}t\,{}^{m}Y + {}^{l}t\,{}^{l}Y + {}^{p}t\,{}^{p}Y)} \frac{\tan(\kappa)}{\kappa} \quad . \tag{15}$$

Using the definition of the magnetoelectric coefficient (8), taking into account the fact that the voltage induced between the electrodes is $U = {}^{p}E_x\,{}^{p}t$. Hence we obtain the expression for the ME voltage coefficient

$$\alpha_E = \frac{{}^{p}Y\,{}^{p}d_{\mathrm{x,zz}}\,{}^{m}q_{\mathrm{z,zz}}}{{}^{p}\varepsilon_{xx}({}^{m}t\,{}^{m}Y + {}^{l}t\,{}^{l}Y + {}^{p}t\,{}^{p}Y)} \frac{{}^{m}Y\,{}^{m}t}{\Delta} \frac{\tan(\kappa)}{\kappa} \tag{16}$$

The results of experimental measurements of the frequency dependence of the structure are show in Figure 2.

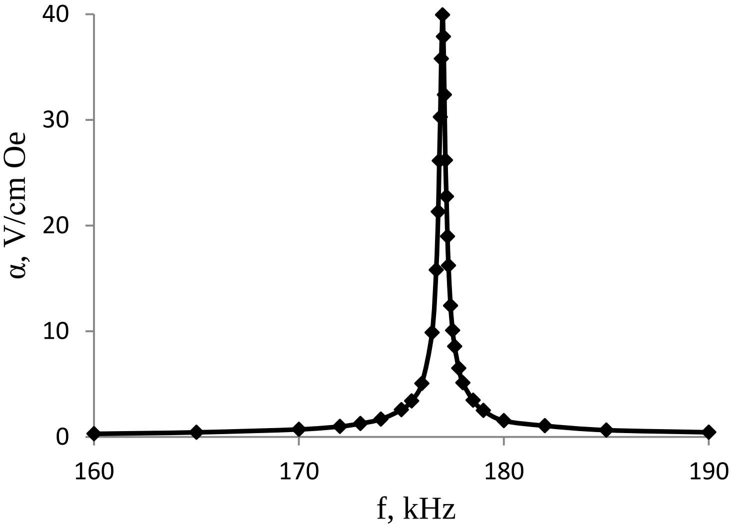

Figure 2. Frequency dependence of the sandwich structure of gallium arsenide-nickel-tin-nickel. The bias field H_{bias} = 360 Oe

As follows from the figure, the frequency dependence has a resonant character. The value of the ME voltage coefficient is somewhat lower than in the pure structure of nickel-gallium arsenide (Filippov *et al.*, 2017). This structure has high the quality factor Q≅700, which is much better than the quality factor of the samples obtained by the gluing method and is comparable to the quality factor of bulk composites.

3. Conclusion

The use of an intermediate tin layer in the electrolytic deposition of nickel on an arsenide-gallium substrate makes it possible to obtain structures with a nickel layer thickness of up to 100 μm. These structures have good adhesion between layers, have good mechanical strength. Such multilayer structures are promising for designing devices based on the ME effect.

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