Technology of production and magnetoelectric characteristics of multilayer structures nickel-tin on the gallium arsenide substrate

D A Filippov¹, I N Manicheva¹, K A Bordashev¹, V M Laletsin², T A Galichyan³

¹Novgorod State University 41, B. St. Peterburgskaya, Velikiy Novgorod, 173003, Russia
²Institute of Technical Acoustics National Academy of Sciences of Belarus 13, Lyudnikava Praspekt, Vitebsk, BY-210023 Belarus
³Institute of Mechanics, National Academy of Sciences of Armenia, 24B Baghramyan Ave., Yerevan, 0019 Armenia

E-mail: Dmitry.Filippov@novsu.ru

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.

1. Introduction
Composite magnetostriective-piezoelectric materials attract attention by the fact that the magnitude of the magnetoelectric (ME) effect in them 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 spin-orbital interaction, the interaction of the electron with the external and intracrystalline electric field [1]. Despite the fact that currently there are dozens of single crystals in which the ME effect is found [2], however, due to its smallness, the ME single crystals have not found wide application in technology. The mechanism of the ME effect in composites is the mechanical interaction of magnetostriective (MS) and piezoelectric (PE) phases. An alternating magnetic field causes mechanical deformations in the MS component, which are transmitted across the interface to the PE component, which leads to a change in polarization and the appearance of electrical voltage. Composite materials can be divided into groups: bulk and layered composites. Bulk composites are mechanically linked mixtures of MS and PE phase powders [3]. Layered composites are structures consisting of alternating layers of magnetic and piezoelectric [4]. Layered structures have several advantages compared to bulk composites: easily polarized, are small in leakage currents [5], as the MS phase, it's possible to use metals having a large coefficient of magnetostriction [6-8]. At the same time, they have a number of disadvantages due to the boundary between the MS and the PE phases. Most layered me structures are obtained by gluing, which leads to a decrease in the quality of the structure, the weakening of the ME effect, undesirable high temperature dependence. In [9,10] 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 [11] the maximum of the ME effect is observed under the condition of equality $p t Y \approx m t Y$, 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 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 [12]. 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. In the production of the multilayer structure, electrolytic deposition was used alternately in the sulfuric acid electrolyte of nickel, with a cathode current density of 1 A/dm² and an electrolyte temperature of 55-65 °C, and then electrodeposition in the tin electrolyte at room temperature and a cathode current density of 2 A/dm². 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 (Figure 1).
At first, the field dependence of the low-frequency ME signal was investigated. At a constant value of the strength of the alternating magnetic field $H=1 \text{ Oe}$, 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 [11]. 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 [13]. 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:

$$
\bar{\rho} \frac{\partial^2 u}{\partial t^2} = \frac{\partial T_z}{\partial z} ,
$$

where $\bar{\rho}$ is the mean value of the sample density, and $T_z$ is the mean value of the stress tensor in the sample. The equations for components of the strain tensor of piezoelectric $\varepsilon S_{zz}$ and magnetic $\mu S_{zz}$, and electric induction $\varepsilon D_z$ are as follows:
\[ \sigma_{zz}^p = \frac{1}{\rho_Y} \sigma_{zz}^{\rho T} + \rho d_{zz}^p \varepsilon_x^p, \]  
\[ \sigma_{zz}^m = \frac{1}{m_Y} \sigma_{zz}^{m T} + m q_{zz}^m H_x, \]  
\[ \rho D_x = \rho e_{xx}^p \varepsilon_x^p + \rho d_{zz}^p \sigma_{zz}^{\rho T}, \]

where \( \sigma_{zz}^{\rho T}, \sigma_{zz}^{m T} \) are components of the stress tensor in the piezoelectric and magnetostrictive phase; \( \rho_Y, m_Y \) are the Young’s modulus in the piezoelectric phase along the <011> (Z-axis) direction, and the magnetic phase, respectively, \( \rho d_{zz}^p \) is the piezoelectric tensor in the XYZ coordinate system (Figure 1), \( \rho e_{xx}^p \) is the dielectric tensor and \( \rho \varepsilon_x^p \) is the x component of the electric field strength vector, \( m q_{zz}^m \) is the piezomagnetic coefficient, \( m H_x \) is the magnetic field strength.

The components of the piezoelectric tensor \( \rho d_{zz}^p \) in the XYZ coordinate system are related to the components of the piezoelectric tensor \( \rho d_{\alpha\beta} \) in the crystallographic coordinate system by the relation:

\[ d_{zz}^p = d_{i4} \beta_{z2} \beta_{z3}, \]

where \( \beta_{z2}, \beta_{z3} \) 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:

\[ u(z) = A \cos(kz) + B \sin(kz), \]

where \( A \) and \( B \) are the integration constants.

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

\[ \omega = \sqrt{ \left( \frac{m Y'' \rho + \rho Y' + \rho Y' \rho T}{m \rho + \rho' + \rho T} \right) k}, \]

where \( \rho = \frac{m Y'' \rho + \rho Y' + \rho Y' \rho T}{m \rho + \rho' + \rho T} \) 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 \( H \), i.e.

\[ \alpha_E = \langle E \rangle / H, \]

where \( \langle E \rangle = U/(\rho' + \rho T + \rho T') \) 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:

\[ \int_0^{\rho' T_{zz}} (\pm L/2, x) dx + \int_{\rho' T_{zz}}^{\rho' T_{zz}} T_{zz}(\pm L/2, x) dx + \int_{\rho' T_{zz}}^{\rho' T_{zz}} T_{zz}(\pm L/2, x) dx = 0 \]

Using these boundary conditions, for integration constants we obtain:

\[ A = 0, \quad \rho = \frac{m Y'' \rho + \rho Y' + \rho H_x}{m \rho + \rho' + \rho T} \]

\[ B = \frac{m Y'' \rho + \rho Y' + \rho H_x}{m \rho + \rho' + \rho T}, \]
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:

\[
\rho D_z = \rho E_z + \rho Y \frac{\partial \rho u}{\partial z} - \rho Y \left( \rho d_z \right)^2 \rho E_z.
\]  

(11)

To determine the electric field \( \rho E_z \) induced in a piezoelectric, we use of the open-circuit condition:

\[
I = \iint \frac{\partial D_z}{\partial t} dz dy = 0,
\]

(12)

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

\[
\rho E_z = \frac{\rho Y \rho d_{zz} n q_{zz}}{\rho E_{xx}} \frac{m H_x}{\Delta} \left( m t m Y + t' Y + \rho Y \right) \tan(\kappa),
\]

(13)

where the notation

\[
\Delta = 1 - k^2 \left( \frac{\rho t \rho Y}{(m t m Y + t' Y + \rho Y)} \right) \tan(\kappa).
\]

(14)

Using the definition of the magnetoelectric coefficient (8), taking into account the fact that the voltage induced between the electrodes is \( \rho E_z \rho t \). Hence we obtain the expression for the ME voltage coefficient

\[
\alpha_E = \frac{\rho Y \rho d_{zz} n q_{zz}}{\rho E_{xx}} \frac{m H_x}{\Delta} \left( m t m Y + t' Y + \rho Y \right) \tan(\kappa).
\]

(15)

Equation (15) determines the frequency dependence of the ME voltage coefficient. This dependence is determined by the parameter \( \kappa = kL/2 \), and the wave number \( k \) is determined by the dispersion relation (6). Therefore, changing the composition it is possible to obtain different frequency characteristics. In Figure.2 the results of experimental measurements of the frequency dependence of the ME voltage coefficient for a structure consisting of sixteen layers of nickel on each side of the sample, a total thickness of 100 microns and seventeen layers of tin, a total thickness of 200 microns. The total thickness of the sandwich structure, taking into account the substrate was 1 mm. Layers on gallium arsenide had a smooth, matte surface, without visible defects.
Figure 2. Frequency dependence of the sandwich structure 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 [12]. This structure has high the quality factor $Q \approx 700$, 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.

5. 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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